

QED in Dispersing and Absorbing Media*

Ludwig Knöll, Stefan Scheel, Dirk-Gunnar Welsch

Theoretisch-Physikalisches Institut
Friedrich-Schiller-Universität Jena
Max-Wien-Platz 1, D-07743 Jena, Germany

Abstract

After giving an outline of the quantization scheme based on the microscopic Hopfield model of a dielectric bulk material, we show how the classical phenomenological Maxwell equations of the electromagnetic field in the presence of dielectric matter of given space- and frequency-dependent complex permittivity can be transferred to quantum theory. Including in the theory the interaction of the medium-assisted field with atomic systems, we present both the minimal-coupling Hamiltonian and the multipolar-coupling Hamiltonian in the Coulomb gauge. To illustrate the concept, we discuss the input-output relations of radiation and the transformation of radiation-field quantum states at absorbing four-port devices, and the spontaneous decay of an excited atom near the surface of an absorbing body and in a spherical micro-cavity with intrinsic material losses. Finally, we give an extension of the quantization scheme to other media such as amplifying media, magnetic media, and nonlinear media.

*In *Coherence and Statistics of Photons and Atoms*, edited by J. Peřina (Wiley, New York, 2001), p. 1.

Contents

1	Introduction	2
2	Hopfield model and Fano diagonalization	4
3	The medium-assisted Maxwell field	8
3.1	Classical basic equations	8
3.2	Field quantization	11
4	Atom–Field Interaction	15
4.1	The minimal-coupling Hamiltonian	15
4.2	The multipolar-coupling Hamiltonian	16
5	Input–output coupling	19
5.1	Operator input–output relations	20
5.2	Quantum-state transformation	24
6	Spontaneous decay	30
6.1	Time dependence of the atom–field system	31
6.2	Atom near a dielectric surface	34
6.3	Real-cavity model of spontaneous decay in a dielectric medium	36
6.4	Cavity QED	38
7	Extensions to other media	42
7.1	Amplifying media	42
7.2	Anisotropic media	42
7.3	Magnetic media	43
7.4	Nonlinear media	44
A	The Green tensor	52
A.1	Basic relations	52
B	Commutation relations	56
C	Equations of motion	58

1 Introduction

As is already known from classical optics, the use of instruments in optical experiments needs careful examination with regard to their action on the light under study. Fibres, beam splitters, cavities, spectral filters etc. are familiar examples of optical instruments, which are typically built up by dielectric bodies. In quantum optics an important aspect is the influence of such material bodies on the quantum features of light including its interaction with (microscopic) atomic systems. For example, let us consider a 50%/50% beam splitter oriented at 45° to an incident light beam. In classical optics the beam splitter simply divides the incoming beam into two (apart from a phase shift) equal outgoing parts propagating perpendicular to each other, and with the same scaling factor the classical noise of the incident field is transferred to the two fields in the output channels of the beam splitter. It is intuitively clear that in quantum optics the noise of the vacuum in the unused input port of the beam splitter introduces additional noise in the two output beams and thus the quantum statistics of the output field may differ significantly from that of the input field provided that the input field is prepared in a nonclassical state.

Moreover, when two light beams that are prepared in non-classical states are superimposed by the beam splitter, then the outgoing field is prepared in a nonclassically correlated bipartite state, also called an entangled state. Entanglement as a typical quantum coherence feature plays an important role in quantum communication. Let us assume that two light beams have been prepared in an entangled state. In order to use them, e.g., in quantum teleportation, the beams should be transmitted through optical channels such as fibres. Here the crucial point is to what extent the entanglement can be preserved during the propagation of the beams, because in any real fibre the absorption gives necessarily rise to an entanglement degradation.

From a more theoretical point of view, a very interesting question is that of the Casimir force between material bodies. A crude physical explanation of the Casimir force is that the vacuum energies of two regions spatially separated by the bodies differ due to the presence of the matter, which in a rough approximation can be described simply by a boundary condition on the quantization volume. In fact, this simple model does not take account of the dispersive and absorptive properties of the matter and fails in the high-frequency limit where the matter becomes transparent.

These few examples show that it is necessary to include in the theory the presence of material bodies when considering the quantized radiation field and its interaction with atoms. In principle, material bodies could be included as a part of the matter to which the radiation field is coupled and treated microscopically. However, there is a class of material bodies whose action can be included in the quantum theory exactly, namely dielectric bodies that respond linearly to the electromagnetic field, the response being described in terms of a phenomenologically introduced space-dependent dielectric permittivity. Such a concept has – similar to classical optics – the benefit of being universally valid, because it uses only general physical properties, without the need of involved *ab initio* calculations.

The quantum theory of radiation in the presence of dielectric matter has been

studied over a long period. Quantization of the electromagnetic field in dielectric media with assumed real and frequency-independent permittivity has been treated extensively [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15]. In the same context, dispersive dielectrics have been considered [16, 17, 18, 19, 20, 21, 22, 23] and attempts have been made to extend the concepts to nonlinear media [24, 25, 26, 27]. However, it is well known that the permittivity is a complex function of frequency which has to satisfy the Kramers–Kronig relations, which state that the real part of the permittivity (responsible for dispersion) and the imaginary part (responsible for absorption) are necessarily connected with each other. A consequence of the existence of the imaginary part of the permittivity is that the commonly used mode expansion of the (macroscopic) electromagnetic field fails, at least in frequency intervals where the absorption cannot be disregarded. Obviously, an expansion of the field in terms of damped (non-orthogonal) waves would not be complete. From statistical mechanics it is clear that dissipation is unavoidably connected with the appearance of a random force which gives rise to an additional noise source of the electromagnetic field. Hence, any quantum theory that is based on the assumption of a real permittivity can only be valid for narrow-bandwidth fields far from medium resonances where absorption can safely be disregarded.

For the last years there has been an increasing number of articles dealing with the problem of the formulation of quantum electrodynamics in dielectric media of given complex permittivity satisfying the Kramers–Kronig relations [28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45]. A systematic and quantum-theoretically consistent approach to the problem of the quantization of the radiation field in absorbing bulk dielectrics is given in [30] on the basis of the microscopic Hopfield model of a dielectric [46]. It is based on an explicit Fano-type diagonalization [47] of a Hamiltonian consisting of the electromagnetic field, a (harmonic-oscillator) polarization field representing the dielectric matter, and a continuous set of (harmonic-oscillator) reservoir variables accounting for absorption. The resulting expression for the vector potential can be written in terms of the Green tensor of the classical scattering problem, which, in fact, makes it possible to perform the quantization of the electromagnetic field in the presence of arbitrary dielectric bodies of (phenomenologically) given permittivities [33, 34, 48], without referring to specific microscopic models of the bodies, which were hard to establish for general systems. The concept is based on a source-quantity representation of the electromagnetic field, in which the electromagnetic-field operators are expressed in terms of a continuous set of fundamental bosonic fields via the Green tensor of the classical problem.

Let us give a brief guide to the topics covered. After giving an outline of the quantization scheme based on the microscopic Hopfield model of a dielectric bulk material (Sec. 2), we show how the classical phenomenological Maxwell equations of the electromagnetic field in the presence of dielectric matter of given space- and frequency-dependent permittivity can be transferred to quantum theory (Sec. 3). For this purpose we first summarize the basic properties of the classical Maxwell equations and express the electromagnetic field in terms of the Green tensor and a continuous set of appropriately chosen dynamical field variables (Sec. 3.1). We then

perform the quantization by identifying the dynamical field variables with bosonic fields associated with the elementary excitations of the composed system (Sec. 3.2). Having quantized the electromagnetic field, the question arises of how to include in the theory the interaction of the medium-assisted field with atomic systems (Sec. 4). In order to answer it, we present both the minimal-coupling Hamiltonian (Sec. 4.1) and the multipolar-coupling Hamiltonian (Sec. 4.2) in the Coulomb gauge. To illustrate the basic theoretical concept, we discuss a number of applications such as the input–output relations of radiation (Sec. 5.1) and the transformation of radiation-field quantum states (Sec. 5.2) at absorbing four-port devices, and the spontaneous decay of an excited atom near the (planar) surface of an absorbing body (Sec. 6.2) and in a (spherical) micro-cavity with intrinsic material losses (Secs. 6.3 and 6.4). For the sake of transparency, we perform the calculations for isotropic media and outline the extension to other media in a separate section at the end (Sec. 7).

2 Hopfield model and Fano diagonalization

Following the quantization scheme in [30], we consider a Hopfield model [46] of a bulk dielectric in which N harmonic oscillator fields describing the polarization of the dielectric medium are linearly coupled to a continuum of harmonic oscillators standing for a reservoir. Such a model leads to an energy flow essentially only in one direction, namely from the medium to the reservoir where it “disappears”, hence it is absorbed. The overall system that consists of the radiation, the dielectric-medium polarization, the reservoir, and couplings between them may be regarded as being a Hamiltonian system whose Lagrangian reads

$$L = \int d^3\mathbf{r} \mathcal{L} = \int d^3\mathbf{r} (\mathcal{L}_{\text{rad}} + \mathcal{L}_{\text{mat}} + \mathcal{L}_{\text{int}}), \quad (2.1)$$

where

$$\mathcal{L}_{\text{rad}} = \frac{1}{2}\varepsilon_0 [(\dot{\mathbf{A}} + \nabla U)^2 - c^2(\nabla \times \mathbf{A})^2] \quad (2.2)$$

(U , scalar potential; \mathbf{A} , vector potential; $c^{-2} = \varepsilon_0\mu_0$), and

$$\mathcal{L}_{\text{mat}} = \sum_{i=1}^N \frac{1}{2}\mu (\dot{\mathbf{X}}_i^2 - \omega_i^2 \mathbf{X}_i^2) + \int_0^\infty d\omega \frac{1}{2}\mu [\dot{\mathbf{X}}^2(\omega) - \omega^2 \mathbf{X}^2(\omega)], \quad (2.3)$$

$$\mathcal{L}_{\text{int}} = - \sum_{i=1}^N \alpha_i (\mathbf{A} \dot{\mathbf{X}}_i + U \nabla \mathbf{X}_i) - \int_0^\infty d\omega \dot{\mathbf{X}}(\omega) \sum_{i=1}^N v_i(\omega) \mathbf{X}_i. \quad (2.4)$$

Here \mathcal{L}_{rad} and \mathcal{L}_{mat} are respectively the free Lagrangian densities of the radiation field¹ and the matter fields [i.e., the medium oscillator fields \mathbf{X}_i and the reservoir oscillator fields $\mathbf{X}(\omega)$ with density μ], and \mathcal{L}_{int} is the interacting part, where the medium–field coupling constants α_i play the role of (electric) polarizabilities, and

¹Note that the vector potential fulfills the Coulomb-gauge condition $\nabla \mathbf{A} = 0$.

the medium–reservoir coupling constants $v_i(\omega)$ are assumed to be square-integrable functions of ω . Introducing the canonical momenta

$$\mathbf{\Pi} = \frac{\partial \mathcal{L}}{\partial \dot{\mathbf{A}}} = \varepsilon_0 \dot{\mathbf{A}}, \quad (2.5)$$

$$\mathbf{Q}_i = \frac{\partial \mathcal{L}}{\partial \dot{\mathbf{X}}_i} = \mu \dot{\mathbf{X}}_i - \alpha_i \mathbf{A}, \quad (2.6)$$

$$\mathbf{Q}(\omega) = \frac{\partial \mathcal{L}}{\partial \dot{\mathbf{X}}(\omega)} = \mu \dot{\mathbf{X}}(\omega) - \sum_{i=1}^N v_i(\omega) \mathbf{X}_i, \quad (2.7)$$

it is not difficult to perform the Legendre transformation and construct the Hamiltonian $H = H_{\text{rad}} + H_{\text{mat}} + H_{\text{int}}$ of the overall system.

Since the dielectric medium is assumed to be infinitely extended, it is convenient to go to the reciprocal space²,

$$\mathbf{A}(\mathbf{r}) \rightarrow \tilde{\mathbf{A}}(\mathbf{k}) = \sum_{\lambda=1}^2 A_{\lambda}(\mathbf{k}) \mathbf{e}_{\lambda}(\mathbf{k}), \quad (2.8)$$

$$\mathbf{X}_i(\mathbf{r}) \rightarrow \tilde{\mathbf{X}}_i(\mathbf{k}) = X_{i\parallel}(\mathbf{k}) \boldsymbol{\kappa} + \sum_{\lambda=1}^2 X_{i\lambda}(\mathbf{k}) \mathbf{e}_{\lambda}(\mathbf{k}) \quad (2.9)$$

$[\boldsymbol{\kappa} = \mathbf{k}/|\mathbf{k}|, \mathbf{e}_{\lambda}(\mathbf{k}) \perp \mathbf{k}]$, and $\tilde{\mathbf{\Pi}}(\mathbf{k})$, $\tilde{\mathbf{Q}}_i(\mathbf{k})$, $\tilde{\mathbf{X}}(\mathbf{k}, \omega)$, $\tilde{\mathbf{Q}}(\mathbf{k}, \omega)$ accordingly, and to introduce, with regard to quantization, the new variables

$$a_{\lambda}(\mathbf{k}) = \sqrt{\frac{\varepsilon_0}{2\hbar\tilde{k}c}} \left[\tilde{k}c A_{\lambda}(\mathbf{k}) + \frac{i}{\varepsilon_0} \Pi_{\lambda}(\mathbf{k}) \right], \quad (2.10)$$

$$b_{i\lambda}(\mathbf{k}) = \sqrt{\frac{\mu}{2\hbar\tilde{\omega}_i}} \left[\tilde{\omega}_i X_{i\lambda}(\mathbf{k}) + \frac{i}{\mu} Q_{i\lambda}(\mathbf{k}) \right], \quad (2.11)$$

$$b_{\lambda}(\mathbf{k}, \omega) = \sqrt{\frac{\mu}{2\hbar\omega}} \left[-i\omega X_{\lambda}(\mathbf{k}, \omega) + \frac{1}{\mu} Q_{\lambda}(\mathbf{k}, \omega) \right], \quad (2.12)$$

with

$$\tilde{k}^2 = k^2 + \sum_{i=1}^N \frac{\alpha_i^2}{\mu \varepsilon_0 c^2}, \quad \tilde{\omega}_i^2 = \omega_i^2 + \int_0^{\infty} d\omega \frac{v_i^2(\omega)}{\mu^2}, \quad (2.13)$$

and

$$b_{i\parallel}(\mathbf{k}) = \sqrt{\frac{\mu}{2\hbar\tilde{\omega}_{i\parallel}}} \left[\tilde{\omega}_{i\parallel} X_{i\parallel}(\mathbf{k}) + \frac{i}{\mu} Q_{i\parallel}(\mathbf{k}) \right], \quad (2.14)$$

$$b_{\parallel}(\mathbf{k}, \omega) = \sqrt{\frac{\mu}{2\hbar\omega}} \left[-i\omega X_{\parallel}(\mathbf{k}, \omega) + \frac{1}{\mu} Q_{\parallel}(\mathbf{k}, \omega) \right] \quad (2.15)$$

²The spatial Fourier transform $\tilde{F}(\mathbf{k})$ of a function $F(\mathbf{r})$ is defined according to the relation $F(\mathbf{r}) = (2\pi)^{-3/2} \int d^3\mathbf{k} \tilde{F}(\mathbf{k}) e^{i\mathbf{k}\mathbf{r}}$.

$$[\tilde{\omega}_{i\parallel}^2 = \tilde{\omega}_i^2 + \alpha_i^2/(\mu\epsilon_0)].$$

The fields are now quantized in the familiar way by conversion of the complex amplitudes $a_\lambda(\mathbf{k})$, $b_{i\lambda(\parallel)}(\mathbf{k})$, and $b_{\lambda(\parallel)}(\mathbf{k}, \omega)$ into bosonic annihilation operators $\hat{a}_\lambda(\mathbf{k})$, $\hat{b}_{i\lambda(\parallel)}(\mathbf{k})$, and $\hat{b}_{\lambda(\parallel)}(\mathbf{k}, \omega)$, and the Hamiltonian can be expressed in terms of the annihilation and creation operators as follows:

$$\hat{H} = \hat{H}^\perp + \hat{H}_{\text{mat}}^\parallel, \quad \hat{H}^\perp = \hat{H}_{\text{rad}} + \hat{H}_{\text{mat}}^\perp + \hat{H}_{\text{int}}^\perp, \quad (2.16)$$

$$\begin{aligned} \hat{H}_{\text{mat}}^\parallel = & \int d^3\mathbf{k} \left\{ \int_0^\infty d\omega \hbar\omega \hat{b}_{\parallel}^\dagger(\mathbf{k}, \omega) \hat{b}_{\parallel}(\mathbf{k}, \omega) + \sum_{i=1}^N \hbar\tilde{\omega}_{i\parallel} \hat{b}_{i\parallel}^\dagger(\mathbf{k}) \hat{b}_{i\parallel}(\mathbf{k}) \right. \\ & \left. + \int_0^\infty d\omega \sum_{i=1}^N \left[\frac{1}{2} \hbar V_i(\omega) \left(\hat{b}_{i\parallel}^\dagger(\mathbf{k}) + \hat{b}_{i\parallel}(-\mathbf{k}) \right) \left(\hat{b}_{\parallel}^\dagger(-\mathbf{k}, \omega) + \hat{b}_{\parallel}(\mathbf{k}, \omega) \right) \right] \right\}, \end{aligned} \quad (2.17)$$

$$\hat{H}_{\text{rad}}^\perp = \sum_{\lambda=1}^2 \int d^3\mathbf{k} \hbar\tilde{k}c \hat{a}_\lambda^\dagger(\mathbf{k}) \hat{a}_\lambda(\mathbf{k}), \quad (2.18)$$

$$\begin{aligned} \hat{H}_{\text{mat}}^\perp = & \sum_{\lambda=1}^2 \int d^3\mathbf{k} \left\{ \int_0^\infty d\omega \hbar\omega \hat{b}_\lambda^\dagger(\mathbf{k}, \omega) \hat{b}_\lambda(\mathbf{k}, \omega) + \sum_{i=1}^N \hbar\tilde{\omega}_i \hat{b}_{i\lambda}^\dagger(\mathbf{k}) \hat{b}_{i\lambda}(\mathbf{k}) \right. \\ & + \int_0^\infty d\omega \sum_{i=1}^N \left[\frac{1}{2} \hbar V_i(\omega) \left(\hat{b}_{i\lambda}^\dagger(\mathbf{k}) + \hat{b}_{i\lambda}(-\mathbf{k}) \right) \left(\hat{b}_\lambda^\dagger(-\mathbf{k}, \omega) + \hat{b}_\lambda(\mathbf{k}, \omega) \right) \right. \\ & \left. \left. + \sum_{j \neq i} \frac{1}{2} \hbar V_i(\omega) V_j(\omega) \left(\hat{b}_{i\lambda}^\dagger(\mathbf{k}) + \hat{b}_{i\lambda}(-\mathbf{k}) \right) \left(\hat{b}_{j\lambda}^\dagger(-\mathbf{k}) + \hat{b}_{j\lambda}(\mathbf{k}) \right) \right] \right\}, \end{aligned} \quad (2.19)$$

$$\hat{H}_{\text{int}}^\perp = \sum_{\lambda=1}^2 \sum_{i=1}^N \int d^3\mathbf{k} \frac{1}{2} i \hbar \Lambda_i(k) \left[\hat{a}_\lambda^\dagger(-\mathbf{k}) + \hat{a}_\lambda(\mathbf{k}) \right] \left[\hat{b}_{i\lambda}^\dagger(\mathbf{k}) + \hat{b}_{i\lambda}(-\mathbf{k}) \right], \quad (2.20)$$

where $V_i(\omega) = (v_i(\omega)/\mu)(\omega/\tilde{\omega}_i)^{1/2}$ and $\Lambda_i(k) = [(\tilde{\omega}_i \alpha_i^2)/(\mu c \varepsilon_0 \tilde{k})]^{1/2}$. The bilinear Hamiltonian can be diagonalized (separately for the transverse and longitudinal parts) by applying a Fano-type technique [47] such that³

$$\hat{H}_{\text{mat}}^\parallel = \int d^3\mathbf{k} \int_0^\infty d\omega \hbar\omega \hat{B}_{\parallel}^\dagger(\mathbf{k}, \omega) \hat{B}_{\parallel}(\mathbf{k}, \omega), \quad (2.21)$$

$$\hat{H}^\perp = \sum_{\lambda=1}^2 \int d^3\mathbf{k} \int_0^\infty d\omega \hbar\omega \hat{C}_\lambda^\dagger(\mathbf{k}, \omega) \hat{C}_\lambda(\mathbf{k}, \omega). \quad (2.22)$$

³A different technique for diagonalization is utilized in [41, 42], where path-integral quantization of the Lagrangian (2.1) is performed.

Since the derivation of the formulas for expressing the old bosonic operators $\hat{b}_{i\parallel}(\mathbf{k})$, $\hat{b}_{i\parallel}^\dagger(\mathbf{k})$, $\hat{b}_{\parallel}(\mathbf{k}, \omega)$, $\hat{b}_{\parallel}^\dagger(\mathbf{k}, \omega)$ and $\hat{a}_{\lambda}(\mathbf{k}, \omega)$, $\hat{a}_{\lambda}^\dagger(\mathbf{k}, \omega)$, $\hat{b}_{i\lambda}(\mathbf{k})$, $\hat{b}_{i\lambda}^\dagger(\mathbf{k})$, $\hat{b}_{\lambda}(\mathbf{k}, \omega)$, $\hat{b}_{\lambda}^\dagger(\mathbf{k}, \omega)$ in terms of the new bosonic operators $\hat{B}_{\parallel}(\mathbf{k}, \omega)$, $\hat{B}_{\parallel}^\dagger(\mathbf{k}, \omega)$ and $\hat{C}_{\lambda}(\mathbf{k}, \omega)$, $\hat{C}_{\lambda}^\dagger(\mathbf{k}, \omega)$, respectively, is rather lengthy, we renounce it here and refer the reader to [30, 49].

The vector potential and the transverse part of the medium polarization can then be expressed in terms of the (polariton-like) operators $\hat{C}_{\lambda}(\mathbf{k}, \omega)$, $\hat{C}_{\lambda}^\dagger(\mathbf{k}, \omega)$ as [30, 34, 48]

$$\hat{\mathbf{A}}(\mathbf{k}) = - \sum_{\lambda=1}^2 \mathbf{e}_{\lambda}(\mathbf{k}) \sqrt{\frac{\hbar}{\pi \varepsilon_0}} \int_0^{\infty} d\omega \omega \sqrt{\varepsilon_I(\omega)} \tilde{G}(\mathbf{k}, \omega) \hat{C}_{\lambda}(\mathbf{k}, \omega) + \text{H.c.} \quad (2.23)$$

and

$$\begin{aligned} \hat{\mathbf{P}}(\mathbf{k}) &= \sum_{\lambda=1}^2 \sum_{i=1}^N \alpha_i \hat{\mathbf{X}}_{i\lambda}(\mathbf{k}) = -i \sum_{\lambda=1}^2 \mathbf{e}_{\lambda}(\mathbf{k}) \sqrt{\frac{\hbar \varepsilon_0}{\pi}} \\ &\times \left[\int_0^{\infty} d\omega \omega^2 [\varepsilon(\omega) - 1] \sqrt{\varepsilon_I(\omega)} \tilde{G}(\mathbf{k}, \omega) \hat{C}_{\lambda}(\mathbf{k}, \omega) + \text{H.c.} \right] + \hat{\mathbf{P}}_N(\mathbf{k}), \end{aligned} \quad (2.24)$$

$$\hat{\mathbf{P}}_N(\mathbf{k}) = \sum_{\lambda=1}^2 \mathbf{e}_{\lambda}(\mathbf{k}) \sqrt{\frac{\hbar \varepsilon_0}{\pi}} \int_0^{\infty} d\omega i \sqrt{\varepsilon_I(\omega)} \hat{C}_{\lambda}(\mathbf{k}, \omega) + \text{H.c.} \quad (2.25)$$

Here, $\varepsilon(\omega) = \varepsilon_R(\omega) + i\varepsilon_I(\omega)$ is the complex (model) permittivity of the medium, and

$$\tilde{G}(\mathbf{k}, \omega) = -\frac{c^2}{\omega^2 \varepsilon(\omega) - k^2 c^2} \quad (2.26)$$

is the Green function of the classical Maxwell equations with that permittivity. From Eq. (2.24) [together with Eq. (2.25)] it is seen that the (transverse) polarization of the medium consists of two qualitatively different terms. Obviously, the first term is the induced polarization, whose frequency components are given by the frequency components of the electric field strength of the radiation multiplied by $\varepsilon_0[\varepsilon(\omega) - 1]$, and the second term is the (noise) polarization, i.e., the fluctuating component of the polarization that is associated with absorption.

Recalling that the longitudinal part of the electric field operator is given by $\varepsilon_0 \hat{\mathbf{E}}_{\parallel}(\mathbf{k}) = -\sum_i \alpha_i \hat{\tilde{X}}_{i\parallel}(\mathbf{k}) \boldsymbol{\kappa}$ and recalling the transversality of the displacement operator $\hat{\mathbf{D}}(\mathbf{k})$, a relation between $\hat{\mathbf{E}}_{\parallel}$ and a longitudinal (noise) polarization defined analogously to Eq. (2.25) [by replacing $\hat{C}_{\lambda}(\mathbf{k}, \omega)$ with $\hat{B}_{\parallel}(\mathbf{k}, \omega)$] can be derived.

In summary, the quantized electromagnetic field can be expressed, via a source-quantity representation with the classical Green function, in terms of the permittivity and a continuum of harmonic oscillators. It is worth noting that in this formulation there is no explicit hint at the underlying microscopic model. Thus, it seems quite natural to generalize the theory to arbitrary dielectric matter of given space- and frequency-dependent permittivity by transferring the classical source-quantity representation of the electromagnetic field directly to quantum theory.

3 The medium-assisted Maxwell field

From now we will not refer to one or the other microscopic model of the dielectric media. Instead we will start from the familiar phenomenological Maxwell equations, assuming that the permittivity is known, e.g., from measurements. In order to allow for an arbitrary formation of different (non-moving) dielectric bodies in space, we will assume that the permittivity varies with space. For the sake of transparency we will disregard the tensor character of the permittivity, restricting our attention to isotropic media (for the extension to anisotropic media, see Sec. 7.2).

3.1 Classical basic equations

Let us first briefly outline the classical theory and bring it in a form suitable for quantization. The phenomenological Maxwell equations of the electromagnetic field in the presence of dielectric bodies but without additional charge and current densities read

$$\nabla \mathbf{B}(\mathbf{r}) = 0, \quad (3.1)$$

$$\nabla \times \mathbf{E}(\mathbf{r}) + \dot{\mathbf{B}}(\mathbf{r}) = 0, \quad (3.2)$$

$$\nabla \mathbf{D}(\mathbf{r}) = 0, \quad (3.3)$$

$$\nabla \times \mathbf{H}(\mathbf{r}) - \dot{\mathbf{D}}(\mathbf{r}) = 0, \quad (3.4)$$

where the displacement field \mathbf{D} is related to the electric field \mathbf{E} and the polarization field \mathbf{P} according to

$$\mathbf{D}(\mathbf{r}) = \varepsilon_0 \mathbf{E}(\mathbf{r}) + \mathbf{P}(\mathbf{r}), \quad (3.5)$$

and for nonmagnetic matter it may be assumed that

$$\mathbf{H}(\mathbf{r}) = \frac{1}{\mu_0} \mathbf{B}(\mathbf{r}) \quad (3.6)$$

(for the extension to magnetic matter, see Sec. 7.3).

Let us consider arbitrarily inhomogeneous (isotropic) media and assume that the polarization responds linearly and locally to the electric field. In this case, the most general relation between the polarization and the electric field which is in agreement with the causality principle and the dissipation-fluctuation theorem is

$$\mathbf{P}(\mathbf{r}, t) = \varepsilon_0 \int_0^\infty d\tau \chi(\mathbf{r}, \tau) \mathbf{E}(\mathbf{r}, t - \tau) + \mathbf{P}_N(\mathbf{r}, t), \quad (3.7)$$

where $\chi(\mathbf{r}, \tau)$ is the dielectric susceptibility as a function of space and time, and \mathbf{P}_N is the (noise) polarization associated with absorption.

Substitution of this expression into Eq. (3.5) together with Fourier transformation converts this equation to

$$\underline{\mathbf{D}}(\mathbf{r}, \omega) = \varepsilon_0 \varepsilon(\mathbf{r}, \omega) \underline{\mathbf{E}}(\mathbf{r}, \omega) + \underline{\mathbf{P}}_N(\mathbf{r}, \omega), \quad (3.8)$$

and thus

$$\underline{\mathbf{P}}(\mathbf{r}, \omega) = \varepsilon_0 [\varepsilon(\mathbf{r}, \omega) - 1] \underline{\mathbf{E}}(\mathbf{r}, \omega) + \underline{\mathbf{P}}_N(\mathbf{r}, \omega), \quad (3.9)$$

where

$$\varepsilon(\mathbf{r}, \omega) = 1 + \int_0^\infty d\tau \chi(\mathbf{r}, \tau) e^{i\omega\tau} \quad (3.10)$$

is the (relative) permittivity, and the Maxwell equations (3.1) – (3.4) read in the Fourier domain as⁴

$$\nabla \underline{\mathbf{B}}(\mathbf{r}, \omega) = 0, \quad (3.11)$$

$$\nabla \times \underline{\mathbf{E}}(\mathbf{r}, \omega) - i\omega \underline{\mathbf{B}}(\mathbf{r}, \omega) = 0, \quad (3.12)$$

$$\varepsilon_0 \nabla \varepsilon(\mathbf{r}, \omega) \underline{\mathbf{E}}(\mathbf{r}, \omega) = \underline{\rho}_N(\mathbf{r}, \omega), \quad (3.13)$$

$$\nabla \times \underline{\mathbf{B}}(\mathbf{r}, \omega) + i \frac{\omega}{c^2} \varepsilon(\mathbf{r}, \omega) \underline{\mathbf{E}}(\mathbf{r}, \omega) = \mu_0 \underline{\mathbf{j}}_N(\mathbf{r}, \omega). \quad (3.14)$$

Here we have introduced the (noise) charge density

$$\underline{\rho}_N(\mathbf{r}, \omega) = -\nabla \underline{\mathbf{P}}_N(\mathbf{r}, \omega) \quad (3.15)$$

and the (noise) current density

$$\underline{\mathbf{j}}_N(\mathbf{r}, \omega) = -i\omega \underline{\mathbf{P}}_N(\mathbf{r}, \omega), \quad (3.16)$$

which obey the continuity equation

$$\nabla \underline{\mathbf{j}}_N(\mathbf{r}, \omega) - i\omega \underline{\rho}_N(\mathbf{r}, \omega) = 0. \quad (3.17)$$

According to Eq. (3.10), the permittivity $\varepsilon(\mathbf{r}, \omega)$ is a complex function of frequency,

$$\varepsilon(\mathbf{r}, \omega) = \varepsilon_R(\mathbf{r}, \omega) + i \varepsilon_I(\mathbf{r}, \omega). \quad (3.18)$$

The real and imaginary parts, which are responsible for dispersion and absorption, respectively, are uniquely related to each other through the Kramers–Kronig relations

$$\varepsilon_R(\mathbf{r}, \omega) - 1 = \frac{\mathcal{P}}{\pi} \int d\omega' \frac{\varepsilon_I(\mathbf{r}, \omega')}{\omega' - \omega}, \quad (3.19)$$

$$\varepsilon_I(\mathbf{r}, \omega) = -\frac{\mathcal{P}}{\pi} \int d\omega' \frac{\varepsilon_R(\mathbf{r}, \omega') - 1}{\omega' - \omega} \quad (3.20)$$

(\mathcal{P} , principal value). Further, $\varepsilon(\mathbf{r}, \omega)$ as a function of complex ω satisfies the relation

$$\varepsilon(\mathbf{r}, -\omega^*) = \varepsilon^*(\mathbf{r}, \omega) \quad (3.21)$$

and is holomorphic in the upper complex half-plane without zeros. In particular, it approaches unity in the high-frequency limit, i.e., $\varepsilon(\mathbf{r}, \omega) \rightarrow 1$ if $|\omega| \rightarrow \infty$ [50, 51].

⁴Here and in the following the Fourier transform $\underline{F}(\omega)$ of a real function $F(t)$ is defined according to the relation $F(t) = F^{(+)}(t) + F^{(-)}(t)$, where $F^{(+)}(t) = \int_0^\infty d\omega \underline{F}(\omega) e^{-i\omega t}$ and $F^{(-)}(t) = [F^{(+)}(t)]^*$.

The Maxwell equations (3.12) and (3.14) imply that $\underline{\mathbf{E}}(\mathbf{r}, \omega)$ obeys the partial differential equation

$$\nabla \times \nabla \times \underline{\mathbf{E}}(\mathbf{r}, \omega) - \frac{\omega^2}{c^2} \varepsilon(\mathbf{r}, \omega) \underline{\mathbf{E}}(\mathbf{r}, \omega) = i\omega\mu_0 \underline{\mathbf{j}}_N(\mathbf{r}, \omega), \quad (3.22)$$

the solution of which can be represented in the form

$$\underline{\mathbf{E}}(\mathbf{r}, \omega) = i\omega\mu_0 \int d^3\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \underline{\mathbf{j}}_N(\mathbf{r}', \omega), \quad (3.23)$$

where the Green tensor $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ has to be determined from the equation

$$\nabla \times \nabla \times \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2}{c^2} \varepsilon(\mathbf{r}, \omega) \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') \quad (3.24)$$

together with the boundary condition at infinity. In Cartesian coordinates, Eq.(3.24) reads

$$\left[(\partial_i^r \partial_k^r - \delta_{ik} \Delta^r) - \delta_{ik} \frac{\omega^2}{c^2} \varepsilon(\mathbf{r}, \omega) \right] G_{kj}(\mathbf{r}, \mathbf{r}', \omega) = \delta_{ij} \delta(\mathbf{r} - \mathbf{r}') \quad (3.25)$$

($\partial_i^r = \partial/\partial x_i$), where over repeated vector-component indices is summed. The Green tensor has the properties that

$$G_{ij}^*(\mathbf{r}, \mathbf{r}', \omega) = G_{ij}(\mathbf{r}, \mathbf{r}', -\omega^*), \quad (3.26)$$

$$G_{ji}(\mathbf{r}', \mathbf{r}, \omega) = G_{ij}(\mathbf{r}, \mathbf{r}', \omega), \quad (3.27)$$

and

$$\int d^3\mathbf{s} \frac{\omega^2}{c^2} \varepsilon_I(\mathbf{s}, \omega) G_{ik}(\mathbf{r}, \mathbf{s}, \omega) G_{jk}^*(\mathbf{r}', \mathbf{s}, \omega) = \text{Im } G_{ij}(\mathbf{r}, \mathbf{r}', \omega). \quad (3.28)$$

The property (3.26) is a direct consequence of the corresponding relation (3.21) for the permittivity. The reciprocity relation (3.27) and the integral relation (3.28) are proven in Appendix A.

The Fourier components of the magnetic induction, $\underline{\mathbf{B}}(\mathbf{r}, \omega)$, and the displacement field, $\underline{\mathbf{D}}(\mathbf{r}, \omega)$, are directly related to the Fourier components of the electric field, $\underline{\mathbf{E}}(\mathbf{r}, \omega)$,

$$\underline{\mathbf{B}}(\mathbf{r}, \omega) = (i\omega)^{-1} \nabla \times \underline{\mathbf{E}}(\mathbf{r}, \omega), \quad (3.29)$$

$$\underline{\mathbf{D}}(\mathbf{r}, \omega) = (\mu_0 \omega^2)^{-1} \nabla \times \nabla \times \underline{\mathbf{E}}(\mathbf{r}, \omega) \quad (3.30)$$

[see Eqs. (3.12), (3.8), (3.16), and (3.22)], and $\underline{\mathbf{E}}(\mathbf{r}, \omega)$ is determined, according to Eq. (3.23), by $\underline{\mathbf{j}}_N(\mathbf{r}, \omega)$. The continuous set of (complex) fields $\underline{\mathbf{j}}_N(\mathbf{r}, \omega)$ [or equivalently, $\underline{\mathbf{P}}_N(\mathbf{r}, \omega)$] can therefore be regarded as playing the role of the set of dynamical variables of the overall system composed of the electromagnetic field and the medium (including the dissipative system). For the following it is convenient to split off some factor from $\underline{\mathbf{P}}_N(\mathbf{r}, \omega)$ and to define the fundamental dynamical variables $\mathbf{f}(\mathbf{r}, \omega)$ according to

$$\underline{\mathbf{P}}_N(\mathbf{r}, \omega) = i \sqrt{\frac{\hbar \varepsilon_0}{\pi}} \varepsilon_I(\mathbf{r}, \omega) \mathbf{f}(\mathbf{r}, \omega). \quad (3.31)$$

3.2 Field quantization

The transition from classical to quantum theory now consists in the replacement of the classical fields $\mathbf{f}(\mathbf{r}, \omega)$ and $\mathbf{f}^*(\mathbf{r}, \omega)$ by the operator-valued bosonic fields $\hat{\mathbf{f}}(\mathbf{r}, \omega)$ and $\hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega)$, respectively, which are associated with the elementary excitations of the composed system within the framework of linear light-matter interaction. Thus the commutation relations are

$$[\hat{f}_k(\mathbf{r}, \omega), \hat{f}_{k'}^\dagger(\mathbf{r}', \omega')] = \delta_{kk'} \delta(\mathbf{r} - \mathbf{r}') \delta(\omega - \omega'), \quad (3.32)$$

$$[\hat{f}_k(\mathbf{r}, \omega), \hat{f}_{k'}(\mathbf{r}', \omega')] = 0, \quad (3.33)$$

and the Hamiltonian of the composed system is

$$\hat{H} = \int d^3\mathbf{r} \int_0^\infty d\omega \hbar \omega \hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega) \hat{\mathbf{f}}(\mathbf{r}, \omega). \quad (3.34)$$

Replacing $\underline{\mathbf{E}}(\mathbf{r}, \omega)$ [Eq. (3.23)], $\underline{\mathbf{B}}(\mathbf{r}, \omega)$ [Eq. (3.29)], and $\underline{\mathbf{D}}(\mathbf{r}, \omega)$ [Eqs. (3.8), (3.30)] by the quantum-mechanical operators, we find, on recalling Eqs. (3.16) and (3.31), that

$$\hat{\underline{\mathbf{E}}}(\mathbf{r}, \omega) = i \sqrt{\frac{\hbar}{\pi \varepsilon_0}} \frac{\omega^2}{c^2} \int d^3\mathbf{r}' \sqrt{\varepsilon_I(\mathbf{r}', \omega)} \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \hat{\mathbf{f}}(\mathbf{r}', \omega), \quad (3.35)$$

$$\hat{\underline{\mathbf{B}}}(\mathbf{r}, \omega) = (i\omega)^{-1} \nabla \times \hat{\underline{\mathbf{E}}}(\mathbf{r}, \omega), \quad (3.36)$$

and

$$\begin{aligned} \hat{\underline{\mathbf{D}}}(\mathbf{r}, \omega) &= \varepsilon_0 \varepsilon(\mathbf{r}, \omega) \hat{\underline{\mathbf{E}}}(\mathbf{r}, \omega) + \hat{\underline{\mathbf{P}}}_N(\mathbf{r}, \omega) \\ &= (\mu_0 \omega^2)^{-1} \nabla \times \nabla \times \hat{\underline{\mathbf{E}}}(\mathbf{r}, \omega), \end{aligned} \quad (3.37)$$

from which the electromagnetic field operators in the Schrödinger picture are obtained by integration over ω :

$$\hat{\mathbf{E}}(\mathbf{r}) = \int_0^\infty d\omega \hat{\underline{\mathbf{E}}}(\mathbf{r}, \omega) + \text{H.c.}, \quad (3.38)$$

$$\hat{\mathbf{B}}(\mathbf{r}) = \int_0^\infty d\omega \hat{\underline{\mathbf{B}}}(\mathbf{r}, \omega) + \text{H.c.}, \quad (3.39)$$

and⁵

$$\hat{\mathbf{D}}(\mathbf{r}) = \hat{\mathbf{D}}^\perp(\mathbf{r}) = \int_0^\infty d\omega \hat{\underline{\mathbf{D}}}(\mathbf{r}, \omega) + \text{H.c.} \quad (3.40)$$

In this way, the electromagnetic field is expressed in terms of the classical Green tensor $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ satisfying the generalized Helmholtz equation (3.24) and the continuum of the fundamental bosonic field variables $\hat{\mathbf{f}}(\mathbf{r}, \omega)$ [and $\hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega)$]. All the information

⁵Here the longitudinal (\mathbf{F}^\parallel) and transverse (\mathbf{F}^\perp) parts of a vector field \mathbf{F} are defined by $\mathbf{F}^{\parallel(\perp)}(\mathbf{r}) = \int d^3\mathbf{r}' \delta^{\parallel(\perp)}(\mathbf{r} - \mathbf{r}') \mathbf{F}(\mathbf{r}')$, with $\delta^{\parallel(\perp)}(\mathbf{r})$ and $\delta^{\perp(\perp)}(\mathbf{r})$ being respectively the longitudinal and transverse tensor-valued δ -functions [Eqs. (B.9) and (B.10)]. Note that for bulk material the transverse polarization field $\hat{\mathbf{P}}^\perp = \hat{\mathbf{D}} - \varepsilon_0 \hat{\mathbf{E}}^\perp$, with $\hat{\mathbf{E}}$ and $\hat{\mathbf{D}}$ being respectively given by Eqs. (3.38) and (3.40) together with Eq. (3.35) and (3.37) exactly corresponds to Eq. (2.24) together with Eq. (2.25).

about the dielectric matter (such as its formation in space and its dispersive and absorptive properties) is contained [via the permittivity $\varepsilon(\mathbf{r}, \omega)$] in the Green tensor of the classical problem. Eqs. (3.38) – (3.40), together with Eqs. (3.35) – (3.37), can be considered as the generalization of the familiar mode decomposition.

A similar formalism which also starts from a causal relation between the polarization and the electric field strength is developed in [37, 38]. The auxiliary fields that are introduced there in order to construct a unitary time evolution in an enlarged Hilbert space can be shown to lead essentially to the field variables $\hat{\mathbf{f}}(\mathbf{r}, \omega)$ and $\hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega)$ considered here. Thus, the representation of the electromagnetic field in [37, 38] corresponds to the Green function representation in Eqs. (3.35) – (3.40).

The quantization scheme meets all the basic requirements of quantum electrodynamics. So it can be shown by using very general properties of the permittivity and the Green tensor that the electric and magnetic fields satisfy the correct (equal-time) commutation relations (see Appendix B)

$$[\hat{E}_k(\mathbf{r}), \hat{E}_l(\mathbf{r}')] = 0 = [\hat{B}_k(\mathbf{r}), \hat{B}_l(\mathbf{r}')], \quad (3.41)$$

$$[\varepsilon_0 \hat{E}_k(\mathbf{r}), \hat{B}_l(\mathbf{r}')] = -i\hbar \epsilon_{klm} \partial_m^x \delta(\mathbf{r} - \mathbf{r}'). \quad (3.42)$$

Obviously, the electromagnetic field operators in the Heisenberg picture satisfy the Maxwell equations (3.1) – (3.4), with the time derivative of any operator \hat{Q} being given by

$$\dot{\hat{Q}} = (i\hbar)^{-1} [\hat{Q}, \hat{H}], \quad (3.43)$$

where \hat{H} is the Hamiltonian (3.34).

Let us briefly comment on the (zero-temperature) statistical implications of the quantization scheme. The vacuum expectation value of $\hat{\underline{\mathbf{E}}}(\mathbf{r}, \omega)$ is obviously zero whereas the fluctuation of $\hat{\underline{\mathbf{E}}}(\mathbf{r}, \omega)$ is not. From Eq. (3.35) together with the commutation relations (3.32) and (3.33) we derive, on using the integral relation (3.28),

$$\langle 0 | \hat{\underline{E}}_k(\mathbf{r}, \omega) \hat{\underline{E}}_l^\dagger(\mathbf{r}', \omega') | 0 \rangle = \frac{\hbar \omega^2}{\pi \epsilon_0 c^2} \text{Im } G_{kl}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega'). \quad (3.44)$$

Equation (3.44) reveals that the fluctuation of the electromagnetic field is determined by the imaginary part of the Green tensor – a result that is consistent with the dissipation–fluctuation theorem⁶ [52]. Thus, the quantization scheme respects both the basic requirements of quantum theory (in terms of the correct commutation relations) and statistical physics (in terms of the dissipation–fluctuation theorem).

So far we have considered the electromagnetic field strengths. Instead, scalar ($\hat{\varphi}$) and vector ($\hat{\mathbf{A}}$) potentials can be introduced and expressed in terms of the fundamental bosonic fields $\hat{\mathbf{f}}(\mathbf{r}, \omega)$ and $\hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega)$. In particular, the potentials in the Coulomb gauge are defined by

$$-\nabla \hat{\varphi}(\mathbf{r}) = \hat{\mathbf{E}}^\parallel(\mathbf{r}), \quad (3.45)$$

⁶Note that the Green tensor plays the role of the response function of the electromagnetic field to an external perturbation.

$$\hat{\mathbf{A}}(\mathbf{r}) = \int_0^\infty d\omega \hat{\mathbf{A}}(\mathbf{r}, \omega) + \text{H.c.}, \quad (3.46)$$

where⁷

$$\hat{\mathbf{A}}(\mathbf{r}, \omega) = (i\omega)^{-1} \hat{\mathbf{E}}^\perp(\mathbf{r}, \omega). \quad (3.47)$$

The canonically conjugated momentum field with respect to $\hat{\mathbf{A}}(\mathbf{r})$ is

$$\hat{\mathbf{\Pi}}(\mathbf{r}) = -i\varepsilon_0 \int_0^\infty d\omega \omega \hat{\mathbf{A}}(\mathbf{r}, \omega) + \text{H.c.}, \quad (3.48)$$

and it is not difficult to verify that $\hat{\mathbf{\Pi}} = -\varepsilon_0 \hat{\mathbf{E}}^\perp$, $\nabla \times \hat{\mathbf{A}} = \hat{\mathbf{B}}$, and $-\dot{\hat{\mathbf{A}}} - \nabla \hat{\varphi} = \hat{\mathbf{E}}$. In addition, $\hat{\mathbf{A}}$ and $\hat{\mathbf{\Pi}}$ satisfy the well-known commutation relations (Appendix B)

$$[\hat{A}_k(\mathbf{r}), \hat{A}_{k'}(\mathbf{r}')] = 0 = [\hat{\Pi}_k(\mathbf{r}), \hat{\Pi}_{k'}(\mathbf{r}')], \quad (3.49)$$

$$[\hat{A}_k(\mathbf{r}), \hat{\Pi}_{k'}(\mathbf{r}')] = i\hbar \delta_{kk'}^\perp(\mathbf{r} - \mathbf{r}'). \quad (3.50)$$

3.2.1 One-dimensional systems

Let us illustrate the main features of the concept for linearly polarized radiation propagating in the x direction, which effectively reduces the system to one spatial dimension [$\hat{\mathbf{A}}(\mathbf{r}) \rightarrow \hat{A}_y(x) \equiv \hat{A}(x)$, $\hat{\mathbf{\Pi}}(\mathbf{r}) \rightarrow \hat{\Pi}_y(x) \equiv \hat{\Pi}(x)$, $\hat{\mathbf{f}}(\mathbf{r}, \omega) \rightarrow \hat{f}(x, \omega)$]. According to Eqs. (3.46) and (3.47) together with Eq. (3.35), the operator of the vector potential is

$$\hat{A}(x) = \sqrt{\frac{\hbar}{\pi\varepsilon_0\mathcal{A}}} \int_0^\infty d\omega \int dx' \frac{\omega}{c^2} \sqrt{\varepsilon_I(x', \omega)} G(x, x', \omega) \hat{f}(x', \omega) + \text{H.c.}, \quad (3.51)$$

and $\hat{\Pi}$ accordingly (\mathcal{A} , normalization area perpendicular to the x direction). Here the Green function $G(x, x', \omega)$ satisfies the equation

$$-\frac{\partial^2}{\partial x^2} G(x, x', \omega) - \frac{\omega^2}{c^2} \varepsilon(x, \omega) G(x, x', \omega) = \delta(x - x'). \quad (3.52)$$

In the simplest case when the spatial variation of the permittivity can be disregarded, then the solution of Eq. (3.52) that satisfies the boundary conditions at $|x|, |x'| \rightarrow \infty$ is

$$G(x, x', \omega) = - \left[2i \frac{\omega}{c} n(\omega) \right]^{-1} \exp \left[i \frac{\omega}{c} n(\omega) |x - x'| \right], \quad (3.53)$$

with $n(\omega) = \sqrt{\varepsilon(\omega)} = n_R(\omega) + i n_I(\omega)$ being the complex refractive index of the medium.

⁷Note that for bulk material Eqs. (3.46) and (3.47) together with Eq. (3.35) exactly correspond to Eq. (2.23).

Substituting the Green function (3.53) into Eq. (3.51), we may rewrite the x' -integral to obtain

$$\begin{aligned} \hat{A}(x) = \int_0^\infty d\omega \left\{ \sqrt{\frac{\hbar}{4\pi\epsilon_0 c \omega n_R(\omega) \mathcal{A}}} \frac{n_R(\omega)}{n(\omega)} \right. \\ \left. \times [e^{in_R(\omega)\omega x/c} \hat{a}_+(x, \omega) + e^{-in_R(\omega)\omega x/c} \hat{a}_-(x, \omega)] + \text{H.c.} \right\}, \end{aligned} \quad (3.54)$$

where

$$\hat{a}_\pm(x, \omega) = i\sqrt{2n_I(\omega)\omega/c} e^{\mp n_I(\omega)\omega x/c} \int_{-\infty}^{\pm x} dx' e^{-in(\omega)\omega x'/c} \hat{f}(\pm x', \omega), \quad (3.55)$$

and from Eq. (3.32) it follows that

$$[\hat{a}_\pm(x, \omega), \hat{a}_\pm^\dagger(x', \omega')] = e^{-n_I(\omega)\omega|x-x'|/c} \delta(\omega - \omega'). \quad (3.56)$$

Obviously, the space-dependent operators $\hat{a}_\pm(x, \omega)$ describe the amplitudes of the damped monochromatic waves propagating to the right (subscript $+$) and left (subscript $-$), and from Eq. (3.55) it follows that $\hat{a}_\pm(x, \omega)$ and $\hat{a}_\pm(x', \omega)$ are related by (spatial) quantum Langevin equations [30, 33],

$$\frac{\partial}{\partial x} \hat{a}_\pm(x, \omega) = \mp n_I(\omega) \frac{\omega}{c} \hat{a}_\pm(x, \omega) + \hat{F}_\pm(x, \omega), \quad (3.57)$$

with

$$\hat{F}_\pm(x, \omega) = \pm i\sqrt{2n_I(\omega)\omega/c} e^{\mp in_R(\omega)\omega x/c} \hat{f}(x, \omega) \quad (3.58)$$

being the operator Langevin noise sources. In particular, when $\langle \hat{f}(x'', \omega) \rangle = 0$ for $|x'' - x| < |x - x'|$ and $|x'' - x'| < |x - x'|$, then

$$\langle \hat{a}_\pm(x, \omega) \rangle = \langle \hat{a}_\pm(x', \omega) \rangle \exp[-n_I(\omega)\omega|x - x'|/c], \quad \pm x \mp x' \geq 0 \quad (3.59)$$

for arbitrary $\langle \hat{a}_\pm(x', \omega) \rangle$.

Equation (3.54) is the extension of the familiar mode decomposition to absorbing media. Let us assume that in a frequency interval $\Delta\omega$ the absorption is sufficiently small, so that for a chosen (finite) propagation interval $|x - x'|$ the condition $n_I(\omega)\omega|x - x'|/c \ll 1$ holds. Then the amplitude operators $\hat{a}_\pm(x, \omega)$ can be regarded as being independent of x for that propagation distance and satisfying the ordinary Bose commutation relations, as is seen from Eqs. (3.55) and (3.56). In the chosen frequency and space intervals, Eq. (3.54) exactly reduces to the familiar expression obtained by mode expansion.

4 Atom–Field Interaction

The interaction of the quantized electromagnetic field with atoms placed inside a dielectric-matter configuration or near dielectric bodies can strongly be influenced by the dielectric medium. A well-known example is the dependence of the rate of spontaneous decay of an excited atom on the properties of a dielectric environment (Sec. 6). In order to study such and related phenomena, the Hamiltonian (3.34) must be supplemented with the Hamiltonian of additional charged particles and their interaction energy with the medium-assisted electromagnetic field.

4.1 The minimal-coupling Hamiltonian

Applying the minimal-coupling scheme, we may write the total Hamiltonian in the form⁸

$$\begin{aligned} \hat{H} = \int d^3\mathbf{r} \int_0^\infty d\omega \, \hbar\omega \, \hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega) \hat{\mathbf{f}}(\mathbf{r}, \omega) + \sum_\alpha \frac{1}{2m_\alpha} \left[\hat{\mathbf{p}}_\alpha - q_\alpha \hat{\mathbf{A}}(\hat{\mathbf{r}}_\alpha) \right]^2 \\ + \frac{1}{2} \int d^3\mathbf{r} \, \hat{\rho}_A(\mathbf{r}) \hat{\varphi}_A(\mathbf{r}) + \int d^3\mathbf{r} \, \hat{\rho}_A(\mathbf{r}) \hat{\varphi}_M(\mathbf{r}), \end{aligned} \quad (4.1)$$

where $\hat{\mathbf{r}}_\alpha$ is the position operator and $\hat{\mathbf{p}}_\alpha$ is the canonical momentum operator of the α th (nonrelativistic) particle of charge q_α and mass m_α . The Hamiltonian (4.1) consists of four terms. The first term is the energy of the electromagnetic field and the medium (including the dissipative system), as introduced in Eq. (3.34). The second term is the kinetic energy of the charged particles, and the third term is their Coulomb energy, where the corresponding scalar potential $\hat{\varphi}_A$ is given by

$$\hat{\varphi}_A(\mathbf{r}) = \int d^3\mathbf{r}' \frac{\hat{\rho}_A(\mathbf{r}')}{4\pi\epsilon_0|\mathbf{r} - \mathbf{r}'|}, \quad (4.2)$$

with

$$\hat{\rho}_A(\mathbf{r}) = \sum_\alpha q_\alpha \delta(\mathbf{r} - \hat{\mathbf{r}}_\alpha) \quad (4.3)$$

being the charge density of the particles. The last term is the Coulomb energy of interaction of the particles with the medium. From Eq. (4.1) it follows that the interaction Hamiltonian reads

$$\hat{H}_{\text{int}} = - \sum_\alpha \frac{q_\alpha}{m_\alpha} \left[\hat{\mathbf{p}}_\alpha - \frac{1}{2} q_\alpha \hat{\mathbf{A}}(\hat{\mathbf{r}}_\alpha) \right] \hat{\mathbf{A}}(\hat{\mathbf{r}}_\alpha) + \int d^3\mathbf{r} \, \hat{\rho}_A(\mathbf{r}) \hat{\varphi}_M(\mathbf{r}). \quad (4.4)$$

Note that in Eqs. (4.1) and (4.4), the vector potential $\hat{\mathbf{A}}$ and the scalar potential $\hat{\varphi}_M$, respectively, must be thought of as being expressed, on using Eq. (3.46) [together with Eqs. (3.47) and (3.35)] and Eq. (3.45) [together with Eqs. (3.38) and (3.35)], in terms of the fundamental fields $\hat{\mathbf{f}}(\mathbf{r}, \omega)$ [and $\hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega)$].

⁸Here and in the following the subscripts A and M are introduced in order to distinguish between atom- and medium-assisted quantities.

In a straightforward but somewhat lengthy calculation (for an example, see Appendix C) it can be shown (by means of the commutation relations derived in Appendix B) that both the operator-valued Maxwell equations

$$\nabla \hat{\mathbf{B}}(\mathbf{r}) = 0, \quad (4.5)$$

$$\nabla \times \hat{\mathbf{E}}(\mathbf{r}) + \dot{\hat{\mathbf{B}}}(\mathbf{r}) = 0, \quad (4.6)$$

$$\nabla \hat{\mathbf{D}}(\mathbf{r}) = \hat{\rho}_A(\mathbf{r}), \quad (4.7)$$

$$\nabla \times \hat{\mathbf{H}}(\mathbf{r}) - \dot{\hat{\mathbf{D}}}(\mathbf{r}) = \hat{\mathbf{j}}_A(\mathbf{r}), \quad (4.8)$$

where

$$\hat{\mathbf{j}}_A(\mathbf{r}) = \frac{1}{2} \sum_{\alpha} q_{\alpha} \left[\dot{\hat{\mathbf{r}}}_{\alpha} \delta(\mathbf{r} - \hat{\mathbf{r}}_{\alpha}) + \delta(\mathbf{r} - \hat{\mathbf{r}}_{\alpha}) \dot{\hat{\mathbf{r}}}_{\alpha} \right], \quad (4.9)$$

and the operator-valued Newtonian equations of motion

$$\dot{\hat{\mathbf{r}}}_{\alpha} = \frac{1}{m_{\alpha}} \left[\hat{\mathbf{p}}_{\alpha} - q_{\alpha} \hat{\mathbf{A}}(\hat{\mathbf{r}}_{\alpha}) \right], \quad (4.10)$$

$$m_{\alpha} \ddot{\hat{\mathbf{r}}}_{\alpha} = q_{\alpha} \left\{ \hat{\mathbf{E}}(\hat{\mathbf{r}}_{\alpha}) + \frac{1}{2} \left[\dot{\hat{\mathbf{r}}}_{\alpha} \times \hat{\mathbf{B}}(\hat{\mathbf{r}}_{\alpha}) - \hat{\mathbf{B}}(\hat{\mathbf{r}}_{\alpha}) \times \dot{\hat{\mathbf{r}}}_{\alpha} \right] \right\} \quad (4.11)$$

are fulfilled. In Eqs. (4.6) – (4.8), the (longitudinal part of the) electric field and the displacement field now contain [compared with Eqs. (3.38) and (3.40)] additional longitudinal components that result from the charge distribution $\hat{\rho}_A(\mathbf{r})$, i.e.,

$$\hat{\mathbf{E}}(\mathbf{r}) = \hat{\mathbf{E}}_M(\mathbf{r}) - \nabla \hat{\varphi}_A(\mathbf{r}) = \left[\int_0^{\infty} d\omega \underline{\hat{\mathbf{E}}}(\mathbf{r}, \omega) + \text{H.c.} \right] - \nabla \hat{\varphi}_A(\mathbf{r}), \quad (4.12)$$

$$\hat{\mathbf{D}}(\mathbf{r}) = \hat{\mathbf{D}}_M(\mathbf{r}) - \varepsilon_0 \nabla \hat{\varphi}_A(\mathbf{r}) = \left[\int_0^{\infty} d\omega \underline{\hat{\mathbf{D}}}(\mathbf{r}, \omega) + \text{H.c.} \right] - \varepsilon_0 \nabla \hat{\varphi}_A(\mathbf{r}), \quad (4.13)$$

with $\underline{\hat{\mathbf{E}}}(\mathbf{r}, \omega)$ and $\underline{\hat{\mathbf{D}}}(\mathbf{r}, \omega)$ being defined by Eqs. (3.35) and (3.37). The Maxwell equations (4.5) and (4.7), respectively, simply follow from the definition of $\hat{\mathbf{B}}(\mathbf{r})$ [Eq. (3.39) together with Eq. (3.36)] and $\hat{\mathbf{D}}(\mathbf{r})$ [Eq. (4.13) together with Eqs. (3.37) and (4.2)]. The Maxwell equations (4.6) and (4.8) are respectively the Heisenberg equations of motion of $\hat{\mathbf{B}}(\mathbf{r})$ and $\hat{\mathbf{D}}(\mathbf{r})$ according to Eq. (3.43), with the Hamiltonian being given by Eq. (4.1), and the Newtonian equations of motion (4.10) and (4.11) are respectively obtained from the Heisenberg equations of motion of $\hat{\mathbf{r}}_{\alpha}$ and $\hat{\mathbf{p}}_{\alpha}$.

4.2 The multipolar-coupling Hamiltonian

In the interaction Hamiltonian (4.4) used in the minimal-coupling scheme the electromagnetic field is expressed in terms of the potentials. With regard to the interaction of the electromagnetic field with (localized) atomic systems (atoms, molecules etc.) the interaction energy is commonly desired to be treated in terms of the field strengths and the atomic polarization and magnetization. This can be achieved by means of a unitary transformation.

Let us consider an atomic system localized at position \mathbf{r}_A and introduce the atomic polarization

$$\hat{\mathbf{P}}_A(\mathbf{r}) = \sum_{\alpha} q_{\alpha} (\hat{\mathbf{r}}_{\alpha} - \mathbf{r}_A) \int_0^1 d\lambda \delta[\mathbf{r} - \mathbf{r}_A - \lambda (\hat{\mathbf{r}}_{\alpha} - \mathbf{r}_A)], \quad (4.14)$$

so that the charge density (4.3) can be rewritten as

$$\hat{\rho}_A(\mathbf{r}) = q_A \delta(\mathbf{r} - \hat{\mathbf{r}}_A) - \nabla \hat{\mathbf{P}}_A(\mathbf{r}), \quad (4.15)$$

with

$$q_A = \sum_{\alpha} q_{\alpha} \quad (4.16)$$

being the total charge of the atomic system. In order to perform the transition from the minimal-coupling scheme to the multipolar-coupling scheme, we apply to the variables the unitary operator

$$\hat{U} = \exp \left[\frac{i}{\hbar} \int d^3\mathbf{r} \hat{\mathbf{P}}_A(\mathbf{r}) \hat{\mathbf{A}}(\mathbf{r}) \right] \quad (4.17)$$

which is known as the Power-Zienau transformation [53, 54, 55, 56]. It is not difficult to prove that the following transformation rules are valid:

$$\hat{\mathbf{r}}'_{\alpha} = \hat{U} \hat{\mathbf{r}}_{\alpha} \hat{U}^{\dagger} = \hat{\mathbf{r}}_{\alpha}, \quad (4.18)$$

$$\begin{aligned} \hat{\mathbf{p}}'_{\alpha} &= \hat{U} \hat{\mathbf{p}}_{\alpha} \hat{U}^{\dagger} \\ &= \hat{\mathbf{p}}_{\alpha} - q_{\alpha} \hat{\mathbf{A}}(\hat{\mathbf{r}}_{\alpha}) - q_{\alpha} \int_0^1 d\lambda \lambda (\hat{\mathbf{r}}_{\alpha} - \mathbf{r}_A) \times \hat{\mathbf{B}}[\mathbf{r}_A + \lambda (\hat{\mathbf{r}}_{\alpha} - \mathbf{r}_A)], \end{aligned} \quad (4.19)$$

$$\begin{aligned} \hat{\mathbf{f}}'(\mathbf{r}, \omega) &= \hat{U} \hat{\mathbf{f}}(\mathbf{r}, \omega) \hat{U}^{\dagger} \\ &= \hat{\mathbf{f}}(\mathbf{r}, \omega) - \frac{i}{\hbar} \sqrt{\frac{\hbar}{\pi \varepsilon_0}} \varepsilon_1(\mathbf{r}, \omega) \frac{\omega}{c^2} \int d^3\mathbf{r}' \hat{\mathbf{P}}_A^{\perp}(\mathbf{r}') \mathbf{G}^*(\mathbf{r}', \mathbf{r}, \omega). \end{aligned} \quad (4.20)$$

Employing equations (4.18) – (4.20) and using Eqs. (3.28), (3.35), and (B.13), we can express the Hamiltonian \hat{H} in Eq. (4.1) in terms of the new variables $\hat{\mathbf{r}}'_{\alpha} = \hat{\mathbf{r}}_{\alpha}$, $\hat{\mathbf{p}}'_{\alpha}$, and $\hat{\mathbf{f}}'(\mathbf{r}, \omega)$ in order to obtain the multipolar Hamiltonian

$$\begin{aligned} \hat{H} &= \int d^3\mathbf{r} \int_0^{\infty} d\omega \hbar \omega \hat{\mathbf{f}}'^{\dagger}(\mathbf{r}, \omega) \hat{\mathbf{f}}'(\mathbf{r}, \omega) \\ &+ \sum_{\alpha} \frac{1}{2m_{\alpha}} \left\{ \hat{\mathbf{p}}'_{\alpha} + q_{\alpha} \int_0^1 d\lambda \lambda (\hat{\mathbf{r}}_{\alpha} - \mathbf{r}_A) \times \hat{\mathbf{B}}'[\mathbf{r}_A + \lambda (\hat{\mathbf{r}}_{\alpha} - \mathbf{r}_A)] \right\}^2 \\ &+ \frac{1}{2} \int d^3\mathbf{r} \hat{\rho}_A(\mathbf{r}) \hat{\varphi}_A(\mathbf{r}) + \frac{1}{2\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_A^{\perp}(\mathbf{r}) \hat{\mathbf{P}}_A^{\perp}(\mathbf{r}) \\ &+ \int d^3\mathbf{r} \hat{\rho}_A(\mathbf{r}) \hat{\varphi}'_M(\mathbf{r}) - \int d^3\mathbf{r} \hat{\mathbf{P}}_A^{\perp}(\mathbf{r}) \hat{\mathbf{E}}_M'^{\perp}(\mathbf{r}), \end{aligned} \quad (4.21)$$

where the relations $\hat{\mathbf{B}}' = \hat{\mathbf{B}}$, $\hat{\varphi}'_{\text{M}} = \hat{\varphi}_{\text{M}}$ [cf. Eq. (B.19)], and

$$\hat{\mathbf{E}}_{\text{M}}'^{\perp}(\mathbf{r}) = \hat{\mathbf{E}}_{\text{M}}^{\perp}(\mathbf{r}) + \frac{1}{\varepsilon_0} \hat{\mathbf{P}}_{\text{A}}^{\perp}(\mathbf{r}) \quad (4.22)$$

are valid.

In particular when the charged particles form a neutral atomic system ($q_{\text{A}} = 0$), then we may write, on integrating by parts and recalling that $\hat{\mathbf{E}}_{\text{A(M)}}^{\parallel} = -\hat{\mathbf{P}}_{\text{A(M)}}^{\parallel}/\varepsilon_0$,

$$\int d^3\mathbf{r} \hat{\rho}_{\text{A}}(\mathbf{r}) \hat{\varphi}_{\text{A}}(\mathbf{r}) = \frac{1}{\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_{\text{A}}^{\parallel}(\mathbf{r}) \hat{\mathbf{P}}_{\text{A}}^{\parallel}(\mathbf{r}), \quad (4.23)$$

and

$$\begin{aligned} \int d^3\mathbf{r} \hat{\rho}_{\text{A}}(\mathbf{r}) \hat{\varphi}_{\text{M}}(\mathbf{r}) &= \frac{1}{\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_{\text{A}}^{\parallel}(\mathbf{r}) \hat{\mathbf{P}}_{\text{M}}^{\parallel}(\mathbf{r}) \\ &= \frac{1}{\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_{\text{A}}(\mathbf{r}) \hat{\mathbf{P}}_{\text{M}}(\mathbf{r}) - \frac{1}{\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_{\text{A}}^{\perp}(\mathbf{r}) \hat{\mathbf{P}}_{\text{M}}^{\perp}(\mathbf{r}). \end{aligned} \quad (4.24)$$

Combining Eqs. (4.21) – (4.24) and taking into account that $\hat{\mathbf{P}}_{\text{M}}' = \hat{\mathbf{P}}_{\text{M}}$ [cf. Eqs. (B.14) and (B.16)], we may rewrite the multipolar Hamiltonian as

$$\begin{aligned} \hat{H} &= \int d^3\mathbf{r} \int_0^{\infty} d\omega \hbar\omega \hat{\mathbf{f}}'^{\dagger}(\mathbf{r}, \omega) \hat{\mathbf{f}}'(\mathbf{r}, \omega) \\ &\quad + \sum_{\alpha} \frac{1}{2m_{\alpha}} \left\{ \hat{\mathbf{p}}'_{\alpha} + q_{\alpha} \int_0^1 d\lambda \lambda (\hat{\mathbf{r}}_{\alpha} - \mathbf{r}_{\text{A}}) \times \hat{\mathbf{B}}'[\mathbf{r}_{\text{A}} + \lambda (\hat{\mathbf{r}}_{\alpha} - \mathbf{r}_{\text{A}})] \right\}^2 \\ &\quad + \frac{1}{2\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_{\text{A}}(\mathbf{r}) \hat{\mathbf{P}}_{\text{A}}(\mathbf{r}) + \frac{1}{\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_{\text{A}}(\mathbf{r}) \hat{\mathbf{P}}_{\text{M}}'(\mathbf{r}) \\ &\quad - \frac{1}{\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_{\text{A}}(\mathbf{r}) \hat{\mathbf{D}}'^{\perp}(\mathbf{r}), \end{aligned} \quad (4.25)$$

where

$$\hat{\mathbf{D}}'^{\perp}(\mathbf{r}) = \hat{\mathbf{D}}_{\text{M}}'^{\perp}(\mathbf{r}) = \varepsilon_0 \hat{\mathbf{E}}_{\text{M}}'^{\perp}(\mathbf{r}) + \hat{\mathbf{P}}_{\text{M}}'^{\perp}(\mathbf{r}). \quad (4.26)$$

From Eq. (4.25) the interaction Hamiltonian is seen to be

$$\begin{aligned} \hat{H}_{\text{int}} &= \frac{1}{\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_{\text{A}}(\mathbf{r}) \hat{\mathbf{P}}_{\text{M}}'(\mathbf{r}) - \frac{1}{\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_{\text{A}}(\mathbf{r}) \hat{\mathbf{D}}'^{\perp}(\mathbf{r}) \\ &\quad - \sum_{\alpha} \frac{q_{\alpha}}{2m_{\alpha}} \int_0^1 d\lambda \lambda \left\{ [(\hat{\mathbf{r}}_{\alpha} - \mathbf{r}_{\text{A}}) \times \hat{\mathbf{p}}'_{\alpha}] \hat{\mathbf{B}}'[\mathbf{r}_{\text{A}} + \lambda (\hat{\mathbf{r}}_{\alpha} - \mathbf{r}_{\text{A}})] + \text{H.c.} \right\} \\ &\quad + \sum_{\alpha} \frac{q_{\alpha}^2}{2m_{\alpha}} \left\{ \int_0^1 d\lambda \lambda (\hat{\mathbf{r}}_{\alpha} - \mathbf{r}_{\text{A}}) \times \hat{\mathbf{B}}'[\mathbf{r}_{\text{A}} + \lambda (\hat{\mathbf{r}}_{\alpha} - \mathbf{r}_{\text{A}})] \right\}^2. \end{aligned} \quad (4.27)$$

The first term on the right-hand side in Eq. (4.27) is a contact term between the medium polarization and the polarization of the atomic system. The second term describes the interaction of the polarization of the atomic system with the transverse

part of the overall displacement field [cf. Eq. (4.26)], and the last two terms refer to magnetic interactions.

So far we have transformed the dynamical variables but left unchanged the Hamiltonian. Instead the Hamiltonian can be transformed to obtain the new one

$$\hat{\mathcal{H}} = \hat{U}^\dagger \hat{H} \hat{U}. \quad (4.28)$$

Obviously, the new Hamiltonian expressed in terms of the new variables formally looks like the untransformed minimal-coupling Hamiltonian expressed in terms of the old variables. Hence expressing in the new Hamiltonian the new variables in terms of the old ones, we arrive at a multipolar-coupling Hamiltonian that (for a neutral atomic system) looks like the Hamiltonian given in Eq. (4.25), i.e.,

$$\begin{aligned} \hat{\mathcal{H}} = & \int d^3\mathbf{r} \int_0^\infty d\omega \hbar\omega \hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega) \hat{\mathbf{f}}(\mathbf{r}, \omega) \\ & + \sum_\alpha \frac{1}{2m_\alpha} \left\{ \hat{\mathbf{p}}_\alpha + q_\alpha \int_0^1 d\lambda \lambda (\hat{\mathbf{r}}_\alpha - \mathbf{r}_A) \times \hat{\mathbf{B}}[\mathbf{r}_A + \lambda (\hat{\mathbf{r}}_\alpha - \mathbf{r}_A)] \right\}^2 \\ & + \frac{1}{2\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_A(\mathbf{r}) \hat{\mathbf{P}}_A(\mathbf{r}) + \frac{1}{\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_A(\mathbf{r}) \hat{\mathbf{P}}_M(\mathbf{r}) \\ & - \frac{1}{\varepsilon_0} \int d^3\mathbf{r} \hat{\mathbf{P}}_A(\mathbf{r}) \hat{\mathbf{D}}^\perp(\mathbf{r}). \end{aligned} \quad (4.29)$$

In fact, the Hamiltonians in Eqs. (4.25) and (4.29) have different meanings. Since the expectation value of an observable associated with an operator \hat{O} must not change, the use of $\hat{\mathcal{H}}$ necessarily requires a transformation of both the operator [$\hat{O} \rightarrow \hat{U}^\dagger \hat{O} \hat{U}$] and the state [$\hat{\varrho} \rightarrow \hat{U}^\dagger \hat{\varrho} \hat{U}$, with $\hat{\varrho}$ being the density operator], so that

$$i\hbar \frac{d}{dt} \langle \hat{O} \rangle = \text{Tr} \left\{ \hat{\varrho} [\hat{O}, \hat{H}] \right\} = \text{Tr} \left\{ \hat{U}^\dagger \hat{\varrho} \hat{U} [\hat{U}^\dagger \hat{O} \hat{U}, \hat{\mathcal{H}}] \right\}. \quad (4.30)$$

Moreover, in Eq. (4.25) the atomic polarization field couples to the transverse component of the overall displacement field, which also contains the transverse component of the atomic polarization, rather than the ordinary transverse displacement field in Eq. (4.29).

5 Input-output coupling

The quantization scheme developed in Sec. 3 is best suited to study the input-output behaviour of optical fields at dielectric devices, because both dispersion and absorption are exactly included in the resulting input-output relations, and characteristic quantities such as the transmission and reflection coefficients of the setup are expressed in terms of its complex refractive-index profile [57]. Input-output relations are a very efficient description of the action of macroscopic bodies on radiation. In particular, they can advantageously be used to obtain the quantum statistics of the

outgoing light from that of the incoming light, either in terms of radiation-field correlation functions or directly in terms of the density matrix. In what follows we will restrict our attention to four-port devices. The extension of the method to (higher-order) multiport-devices is straightforward.

5.1 Operator input-output relations

Let us study the problem of propagation (in the x -direction) of quantized radiation through a dielectric plate that is the middle part of a three-layered planar structure as sketched in Fig. 1. The setup can be characterized by a piecewise constant permittivity

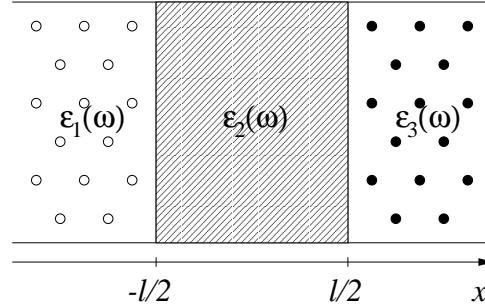


Figure 1: Scheme of a three-layered structure: a dielectric plate of thickness l and permittivity $\epsilon_2(\omega)$ is surrounded by dielectric matter of permittivities $\epsilon_1(\omega)$ (on the left-hand side) and $\epsilon_3(\omega)$ (on the right-hand side).

$$\epsilon(x, \omega) = \sum_{j=1}^3 \lambda_j(x) \epsilon_j(\omega), \quad \lambda_j(x) = \begin{cases} 1 & \text{if } x_{j-1} < x < x_j \\ 0 & \text{otherwise} \end{cases}, \quad (5.1)$$

where $\epsilon_j(\omega)$ is the complex permittivity of the j th domain, and $x_0 \rightarrow -\infty$, $x_1 = -l/2$, $x_2 = l/2$, and $x_3 \rightarrow +\infty$. The Green function $G(x, x', \omega)$ satisfies the partial differential equation (3.52) from which it follows that it can be decomposed into two parts⁹

$$G(x, x', \omega) = \sum_{j=1}^3 \lambda_j(x) \lambda_j(x') G_j^{\text{bulk}}(x, x', \omega) + R(x, x', \omega), \quad (5.2)$$

where $G_j^{\text{bulk}}(x, x', \omega)$ is the Green function of the bulk material as given by Eq. (3.53) (with ϵ_j in place of ϵ), and the reflection term $R(x, x', \omega)$ is a solution to the homogeneous wave equation,

$$R(x, x', \omega) = \sum_{j=1}^3 \lambda_j(x) [C_{j+}(x', \omega) e^{in_j(\omega)\omega x/c} + C_{j-}(x', \omega) e^{-in_j(\omega)\omega x/c}], \quad (5.3)$$

⁹For the calculation of Green tensors and a large variety of examples, see [58].

where the coefficients $C_{j\pm}(x', \omega)$ are to be determined in such a way that continuity and differentiability at the surfaces of discontinuity at $x = -l/2$ and $x = l/2$ are ensured. Combining Eqs. (3.51) and (5.2) [together with Eq. (5.3)], the vector potential $\hat{A}(x)$ for the j th domain may be represented as, similar to Eq. (3.54),

$$\begin{aligned} \hat{A}(x) = \int_0^\infty d\omega \left\{ \sqrt{\frac{\hbar}{4\pi\epsilon_0 c \omega n_{jR}(\omega) \mathcal{A}}} \frac{n_{jR}(\omega)}{n_j(\omega)} \right. \\ \left. \times \left[e^{in_{jR}(\omega)\omega x/c} \hat{a}_{j+}(x, \omega) + e^{-in_{jR}(\omega)\omega x/c} \hat{a}_{j-}(x, \omega) \right] + \text{H.c.} \right\} \end{aligned} \quad (5.4)$$

$[x_{j-1} \leq x \leq x_j]$, where the dependence on x of the amplitude operators $\hat{a}_{j\pm}(x, \omega)$ is governed by quantum Langevin equations of the type given in Eq. (3.57) together with Eq. (3.56).

The amplitude operators of the incoming fields are given according to Eq. (3.55) and satisfy the commutation relations¹⁰

$$[\hat{a}_{1+}(x, \omega), \hat{a}_{1+}^\dagger(x', \omega')] = e^{-n_{1I}(\omega)\omega|x-x'|/c} \delta(\omega - \omega'), \quad (5.5)$$

$$[\hat{a}_{3-}(x, \omega), \hat{a}_{3-}^\dagger(x', \omega')] = e^{-n_{3I}(\omega)\omega|x-x'|/c} \delta(\omega - \omega'), \quad (5.6)$$

$$[\hat{a}_{1+}(x, \omega), \hat{a}_{3-}^\dagger(x', \omega')] = 0 \quad (5.7)$$

[cf. Eq. (3.56)]. Thus, the incoming fields from the left and right behave like the fields in the corresponding bulk dielectrics and may be regarded as independent variables. Further, it can be shown that the amplitude operators of the outgoing fields can be related to the amplitude operators of the incoming fields and appropriately chosen operators $\hat{g}_\pm(\omega)$ of the plate as

$$\begin{pmatrix} \hat{a}_{1-}(-\frac{1}{2}l, \omega) \\ \hat{a}_{3+}(\frac{1}{2}l, \omega) \end{pmatrix} = \mathbf{T}(\omega) \begin{pmatrix} \hat{a}_{1+}(-\frac{1}{2}l, \omega) \\ \hat{a}_{3-}(\frac{1}{2}l, \omega) \end{pmatrix} + \mathbf{A}(\omega) \begin{pmatrix} \hat{g}_+(\omega) \\ \hat{g}_-(\omega) \end{pmatrix}, \quad (5.8)$$

where the 2×2 -matrices $\mathbf{T}(\omega)$ and $\mathbf{A}(\omega)$ are the characteristic transformation and absorption matrices of the plate expressed in terms of the thickness and the permittivity of the plate and the permittivities of the surrounding media, and the operators $\hat{g}_\pm(\omega)$ read

$$\begin{aligned} \hat{g}_\pm(\omega) = i \sqrt{\frac{\omega}{2c\lambda_\pm(l, \omega)}} e^{in_2(\omega)\omega l/(2c)} \\ \times \int_{-l/2}^{l/2} dx' \left[e^{in_2(\omega)\omega x'/c} \pm e^{-in_2(\omega)\omega x'/c} \right] \hat{f}(x', \omega), \end{aligned} \quad (5.9)$$

¹⁰Note that the amplitude operators of the field inside the plate (domain 2) and the amplitude operators of the outgoing fields are not given according to Eq. (3.55) and thus do not satisfy commutation relations of this type in general.

with

$$\lambda_{\pm}(l, \omega) = e^{-n_{2I}(\omega)\omega l/c} \left\{ \frac{\sinh[n_{2I}(\omega)\omega l/c]}{n_{2I}(\omega)} \pm \frac{\sin[n_{2R}(\omega)\omega l/c]}{n_{2R}(\omega)} \right\} \quad (5.10)$$

(for details, see [57]). It is not difficult to prove, on recalling the basic commutation relations (3.32) and (3.33), that the commutation relations

$$[\hat{g}_{\pm}(\omega), \hat{g}_{\pm}^{\dagger}(\omega')] = \delta(\omega - \omega'), \quad (5.11)$$

$$[\hat{g}_{\pm}(\omega), \hat{g}_{\mp}^{\dagger}(\omega')] = 0 \quad (5.12)$$

are valid. Hence, the operators $\hat{g}_{\pm}(\omega)$ and $\hat{g}_{\pm}^{\dagger}(\omega)$ are respectively annihilation and creation operators of bosonic excitations associated with the plate. Obviously, they commute with the amplitude operators of the incoming fields $\hat{a}_{1+}(x, \omega)$ and $\hat{a}_{3-}(x, \omega)$ and are thus independent variables. It should be mentioned that the output amplitude operators $\hat{a}_{1-}(x, \omega)$, $x \leq -l/2$, and $\hat{a}_{3+}(x, \omega)$, $x \geq l/2$, can easily be obtained from $\hat{a}_{1-}(-l/2, \omega)$ and $\hat{a}_{3+}(l/2, \omega)$, respectively, by means of the corresponding solutions of the Langevin equations (3.57).

Whereas the input amplitude operators $\hat{a}_{1+}(-l/2, \omega)$, $\hat{a}_{1+}^{\dagger}(-l/2, \omega)$ and $\hat{a}_{3-}(l/2, \omega)$, $\hat{a}_{3-}^{\dagger}(l/2, \omega)$ satisfy ordinary bosonic commutation relations [see Eqs. (5.5) – (5.7)], the amplitude operators of the outgoing fields, $\hat{a}_{1-}(-l/2, \omega)$, $\hat{a}_{1-}^{\dagger}(-l/2, \omega)$ and $\hat{a}_{3+}(l/2, \omega)$, $\hat{a}_{3+}^{\dagger}(l/2, \omega)$ do not, if the plate is surrounded by (absorbing) matter. When the plate is surrounded by vacuum, then the characteristic transformation and absorption matrices $\mathbf{T}(\omega)$ and $\mathbf{A}(\omega)$ fulfill the matrix relation¹¹

$$\mathbf{T}(\omega)\mathbf{T}^+(\omega) + \mathbf{A}(\omega)\mathbf{A}^+(\omega) = \mathbf{I} \quad (5.13)$$

(\mathbf{I} , unit matrix), and from Eq. (5.8) it then follows that the output amplitude operators also satisfy bosonic commutation relations. In fact, it can be shown that this is not only true at the very input and output ports of the plate but in the whole half-spaces on the left and the right. In this case both the input amplitude operators and the output amplitude operators can be regarded as being bosonic operators associated with ordinary monochromatic incoming and outgoing modes, respectively, and the matrices $\mathbf{T}(\omega)$ and $\mathbf{A}(\omega)$ read as $[n(\omega) \equiv n_2(\omega)]$

$$T_{11}(\omega) = T_{22}(\omega) = e^{-i\omega l/c} r(\omega) [1 - t_1(\omega) e^{2in(\omega)\omega l/c} \vartheta(\omega) t_2(\omega)], \quad (5.14)$$

$$T_{12}(\omega) = T_{21}(\omega) = e^{-i\omega l/c} t_1(\omega) e^{in(\omega)\omega l/c} \vartheta(\omega) t_2(\omega), \quad (5.15)$$

$$\begin{aligned} A_{11}(\omega) = A_{21}(\omega) = & \sqrt{n_I(\omega)n_R(\omega)} e^{-i\omega l/(2c)} t_1(\omega) \vartheta(\omega) \\ & \times \sqrt{\lambda_+(l, \omega)} [1 - e^{in(\omega)\omega l/c} r(\omega)], \end{aligned} \quad (5.16)$$

¹¹Note that a unitary transformation together with rescaling can be applied to the output amplitude operators such that the new operators are bosonic and thus Eq. (5.13) can be assumed to be valid, without loss of generality.

$$A_{12}(\omega) = -A_{22}(\omega) = \sqrt{n_I(\omega)n_R(\omega)}e^{-i\omega l/(2c)}t_1(\omega)\vartheta(\omega) \times \sqrt{\lambda_-(l, \omega)} \left[1 + e^{in(\omega)\omega l/c}r(\omega)\right]. \quad (5.17)$$

Here,

$$r(\omega) = \frac{1 - n(\omega)}{1 + n(\omega)} \quad (5.18)$$

and

$$t_1(\omega) = \frac{2}{1 + n(\omega)}, \quad t_2(\omega) = \frac{2n(\omega)}{1 + n(\omega)} \quad (5.19)$$

are the interface reflection and transmission coefficients, respectively, and the factor

$$\vartheta(\omega) = \left[1 - r^2(\omega)e^{2in(\omega)\omega l/c}\right]^{-1} \quad (5.20)$$

arises from multiple reflections inside the plate.

Operator input-output relations of the type given in Eq. (5.8) are of course valid also for more complicated four-port devices such as multilayer plates. Obviously, the only difference consists in the actual expressions for the characteristic transformation and absorption matrices. For notational reasons it will be convenient to call the input amplitude operators $\hat{a}_i(\omega)$ [i.e., $\hat{a}_{1+}(-l/2, \omega) \rightarrow \hat{a}_1(\omega)$, $\hat{a}_{3-}(l/2, \omega) \rightarrow \hat{a}_2(\omega)$], the output amplitude operators $\hat{b}_i(\omega)$ [i.e., $\hat{a}_{1-}(-l/2, \omega) \rightarrow \hat{b}_1(\omega)$, $\hat{a}_{3+}(l/2, \omega) \rightarrow \hat{b}_2(\omega)$], the device operators $\hat{g}_i(\omega)$ [i.e., $\hat{g}_{\pm}(\omega) \rightarrow \hat{g}_i(\omega)$] (see Fig. 2), and to introduce the definitions

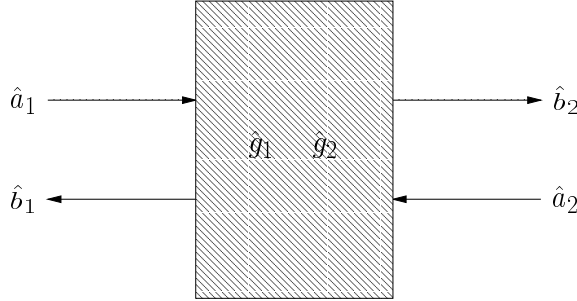


Figure 2: Scheme of a four-port device: Two incoming fields [photonic operators $\hat{a}_1(\omega)$ and $\hat{a}_2(\omega)$] are superimposed to produce two outgoing fields [photonic operators $\hat{b}_1(\omega)$ and $\hat{b}_2(\omega)$], the $\hat{g}_j(\omega)$ being the operators of the relevant device excitations.

$$\hat{\mathbf{a}}(\omega) = \begin{pmatrix} \hat{a}_1(\omega) \\ \hat{a}_2(\omega) \end{pmatrix}, \quad \hat{\mathbf{b}}(\omega) = \begin{pmatrix} \hat{b}_1(\omega) \\ \hat{b}_2(\omega) \end{pmatrix}, \quad \hat{\mathbf{g}}(\omega) = \begin{pmatrix} \hat{g}_1(\omega) \\ \hat{g}_2(\omega) \end{pmatrix}. \quad (5.21)$$

The operator input-output relations (5.8) at a four-port device can then be written in the compact form of

$$\hat{\mathbf{b}}(\omega) = \mathbf{T}(\omega)\hat{\mathbf{a}}(\omega) + \mathbf{A}(\omega)\hat{\mathbf{g}}(\omega). \quad (5.22)$$

In what follows we assume that the characteristic transformation and absorption matrices $\mathbf{T}(\omega)$ and $\mathbf{A}(\omega)$, respectively, obey the equation (5.13).

5.2 Quantum-state transformation

The operator input–output relations (5.22) can be used to calculate various moments and correlations of the outgoing fields in a straightforward way [57, 59]. The output operators are expressed in terms of the input operators and the device operators which for themselves act on the quantum state the incoming field and the device are prepared in. Instead the photonic operators may be left unchanged but the quantum state is transformed. This equivalent procedure is suitable in view of problems, such as the determination of the entanglement of the outgoing fields, where knowledge of the quantum state of the outgoing field as a whole is required. Hence, we are interested in a unitary transformation that transforms the input-state density operator $\hat{\rho}_{\text{in}}$ (i.e., the density operator of the quantum state the incoming field and the device are prepared in) into an output-state density operator $\hat{\rho}_{\text{out}}$ according to

$$\hat{\rho}_{\text{out}} = \hat{U} \hat{\rho}_{\text{in}} \hat{U}^\dagger, \quad (5.23)$$

from which the density operator of the state the outgoing field is prepared in can be obtained by taking the trace with regard to the device variables.

Let us shortly digress to lossless devices and assume, for a moment, that $\mathbf{A}(\omega) = 0$. It is clear from Eq. (5.13) that in this case the characteristic transformation matrix $\mathbf{T}(\omega)$ must be a unitary matrix and thus represents for each ω [and $\det \mathbf{T}(\omega) = 1$] an element of the group $\text{SU}(2)$ [60, 61, 62, 63, 64, 65]. For absorbing devices this can surely not be the case. Because of the coupling to the environment, we will definitely not be able to construct any unitary transformation that acts on the electromagnetic field operators alone. But we may look for one in the larger Hilbert space that comprises both the electromagnetic field and the device. Introducing some auxiliary field operators $\hat{\mathbf{h}}(\omega)$ and defining the “four-vectors”

$$\hat{\boldsymbol{\alpha}}(\omega) = \begin{pmatrix} \hat{\mathbf{a}}(\omega) \\ \hat{\mathbf{g}}(\omega) \end{pmatrix}, \quad \hat{\boldsymbol{\beta}}(\omega) = \begin{pmatrix} \hat{\mathbf{b}}(\omega) \\ \hat{\mathbf{h}}(\omega) \end{pmatrix}, \quad (5.24)$$

we may extend the input–output relations (5.22) to the four-form of

$$\hat{\boldsymbol{\beta}}(\omega) = \boldsymbol{\Lambda}(\omega) \hat{\boldsymbol{\alpha}}(\omega), \quad (5.25)$$

where $\boldsymbol{\Lambda}(\omega)$ is a unitary 4×4 -matrix, hence $\boldsymbol{\Lambda}(\omega) \boldsymbol{\Lambda}^\dagger(\omega) = \mathbf{I}$. After separation of some phases from the matrices $\mathbf{T}(\omega)$ and $\mathbf{A}(\omega)$ and inclusion of them in the input operators, the matrix $\boldsymbol{\Lambda}(\omega)$ can be regarded (for each ω) as an element of the group $\text{SU}(4)$, and it can be expressed in terms of the matrices $\mathbf{T}(\omega)$ and $\mathbf{A}(\omega)$ [66] as

$$\boldsymbol{\Lambda}(\omega) = \begin{pmatrix} \mathbf{T}(\omega) & \mathbf{A}(\omega) \\ -\mathbf{S}(\omega) \mathbf{C}^{-1}(\omega) \mathbf{T}(\omega) & \mathbf{C}(\omega) \mathbf{S}^{-1}(\omega) \mathbf{A}(\omega) \end{pmatrix}, \quad (5.26)$$

where

$$\mathbf{C}(\omega) = \sqrt{\mathbf{T}(\omega) \mathbf{T}^\dagger(\omega)} \quad (5.27)$$

and

$$\mathbf{S}(\omega) = \sqrt{\mathbf{A}(\omega) \mathbf{A}^\dagger(\omega)} \quad (5.28)$$

are commuting positive Hermitian 2×2 -matrices. Note that $\mathbf{C}^2(\omega) + \mathbf{S}^2(\omega) = \mathbf{I}$.

The matrix transformation in Eq. (5.25) may be realized also as a unitary operator transformation

$$\hat{\beta}(\omega) = \hat{U}^\dagger \hat{\alpha}(\omega) \hat{U} = \mathbf{\Lambda}(\omega) \hat{\alpha}(\omega), \quad (5.29)$$

where

$$\hat{U} = \exp \left\{ -i \int_0^\infty d\omega [\hat{\alpha}^\dagger(\omega)]^T \mathbf{\Phi}(\omega) \hat{\alpha}(\omega) \right\}, \quad (5.30)$$

with the 4×4 -matrix $\mathbf{\Phi}(\omega)$ being defined according to

$$\exp[-i\mathbf{\Phi}(\omega)] = \mathbf{\Lambda}(\omega). \quad (5.31)$$

Obviously, \hat{U} is just the unitary operator that transforms $\hat{\varrho}_{\text{in}}$ into $\hat{\varrho}_{\text{out}}$ in Eq. (5.23). Since the input density operator can be regarded as being an operator functional of $\hat{\alpha}(\omega)$ and $\hat{\alpha}^\dagger(\omega)$, $\hat{\varrho}_{\text{in}} = \hat{\varrho}_{\text{in}}[\hat{\alpha}(\omega), \hat{\alpha}^\dagger(\omega)]$, from Eqs. (5.23) and (5.29) it follows that the transformed density operator can be given by

$$\hat{\varrho}_{\text{out}} = \hat{\varrho}_{\text{in}}[\mathbf{\Lambda}^+(\omega) \hat{\alpha}(\omega), \mathbf{\Lambda}^T(\omega) \hat{\alpha}^\dagger(\omega)]. \quad (5.32)$$

Projecting $\hat{\varrho}_{\text{out}}$ onto the Hilbert space of the radiation field then yields the density operator of the outgoing fields

$$\hat{\varrho}_{\text{out}}^{(\text{F})} = \text{Tr}^{(\text{D})} \{ \hat{\varrho}_{\text{in}}[\mathbf{\Lambda}^+(\omega) \hat{\alpha}(\omega), \mathbf{\Lambda}^T(\omega) \hat{\alpha}^\dagger(\omega)] \}, \quad (5.33)$$

where $\text{Tr}^{(\text{D})}$ means the trace with respect to the device.¹²

It is often useful and illustrative to describe quantum states in terms of phase-space functions such as the familiar s -parametrized phase-space functions (see, e.g., [68, 69]). From Eq. (5.32) it follows that the s -parametrized phase-space functional $P_{\text{out}}[\alpha(\omega); s]$ that corresponds to $\hat{\varrho}_{\text{out}}$ is simply given by¹³

$$P_{\text{out}}[\alpha(\omega); s] = P_{\text{in}}[\mathbf{\Lambda}^+(\omega) \alpha(\omega); s], \quad (5.34)$$

so that the phase-space functional of the outgoing radiation reads as

$$P_{\text{out}}^{(\text{F})}[\alpha(\omega); s] = \int \mathcal{D}\mathbf{g} P_{\text{out}}[\alpha(\omega); s] = \int \mathcal{D}\mathbf{g} P_{\text{in}}[\mathbf{\Lambda}^+(\omega) \alpha(\omega); s], \quad (5.35)$$

where the functional integration (notation $\mathcal{D}\mathbf{g}$) is taken over the continua of the complex phase-space variables $g_1(\omega)$ and $g_2(\omega)$ of the dielectric device. In Eq. (5.34) we have used the fact that application of the unitary transformation under consideration

¹²The similarity to the usual open-systems approach to dissipation is not accidental. However, master (or related) equations, to which an open-systems theory would lead [67], are not required here, because the action of the environment (i.e., the device) is explicitly known. For chosen quantum state the device is initially prepared in, all the necessary information is contained in the characteristic transformation and absorption matrices $\mathbf{T}(\omega)$ and $\mathbf{A}(\omega)$, which in turn are determined by the Green function of the phenomenological Maxwell equations of the classical problem.

¹³Since ω is continuous, $P_{\text{in}}[\alpha(\omega); s]$ and $P_{\text{out}}[\alpha(\omega); s]$ are functionals rather than functions.

implies preservation of operator ordering, i.e., the annihilation and creation operators are not mixed by the quantum-state transformation. It should be pointed out that this is not the case for amplifying devices [70].

For practical purposes a description of the incoming and outgoing radiation in terms of discrete (quasi-monochromatic) modes is frequently preferred to be used. For this, we divide the frequency axis into sufficiently small intervals¹⁴ of mid-frequencies ω_m and widths $\Delta\omega_m$ and define the discrete photonic input operators

$$\hat{\alpha}_m = \frac{1}{\sqrt{\Delta\omega_m}} \int_{\Delta\omega_m} d\omega \hat{\alpha}(\omega), \quad (5.36)$$

and the discrete photonic output operators $\hat{\beta}_m$ accordingly. Then we assign to each pair of operators $\hat{\alpha}_m$ and $\hat{\beta}_m$ the input-output relation (5.25) with the 4×4 -matrix $\mathbf{\Lambda}_m = \mathbf{\Lambda}(\omega_m)$, and, according to Eq. (5.30), the unitary operator \hat{U} then reads as

$$\hat{U} = \prod_m \hat{U}_m, \quad (5.37)$$

where

$$\hat{U}_m = \exp \left[-i (\hat{\alpha}_m^\dagger)^T \mathbf{\Phi}_m \hat{\alpha}_m \right] \quad (5.38)$$

with $\mathbf{\Phi}_m = \mathbf{\Phi}(\omega_m)$. In particular, the functional integral in Eq. (5.35) becomes an ordinary multiple integral.

5.2.1 Examples

Let us restrict our attention to (quasi-)monochromatic fields, so that it is sufficient to consider only a single frequency component ω_m . Suppose the incoming field and the device are prepared in coherent states¹⁵

$$|\psi_{\text{in}}\rangle = |\gamma\rangle = \exp(\gamma^T \hat{\alpha}^\dagger - \gamma^+ \hat{\alpha}) |0\rangle, \quad \gamma = \begin{pmatrix} \mathbf{c} \\ \mathbf{d} \end{pmatrix}, \quad (5.39)$$

with c_j and d_j ($j=1, 2$), respectively, being the coherent-state amplitudes of the input fields and the device. Application of Eq. (5.32) yields $|\psi_{\text{out}}\rangle = |\gamma'\rangle$, with $\gamma' = \mathbf{\Lambda}\gamma$ in place of γ in Eq. (5.39), and it follows, on applying Eq. (5.33), that the outgoing fields are prepared in coherent states, i.e.,

$$\hat{\rho}_{\text{out}}^{(\text{F})} = |\mathbf{c}'\rangle \langle \mathbf{c}'|, \quad \mathbf{c}' = \mathbf{T}\mathbf{c} + \mathbf{A}\mathbf{d}. \quad (5.40)$$

Thus, the coherent-state amplitudes c'_1 and c'_2 of the outgoing fields are not only determined by the characteristic transformation matrix \mathbf{T} but also by the absorption matrix \mathbf{A} via the coherent-state amplitudes of the device.

Next let us consider the case where the field in one of the two input channels is prepared in an n -photon Fock state and the field in the other input channel and the

¹⁴The matrices $\mathbf{T}(\omega)$ and $\mathbf{A}(\omega)$ must not change substantially over an interval.

¹⁵For notational convenience we omit the subscript m .

device are left in vacuum, i.e., $|\psi_{\text{in}}\rangle = |n000\rangle$. The Wigner function of the input state reads¹⁶

$$W_{\text{in}}(\boldsymbol{\alpha}) = (-1)^n \left(\frac{2}{\pi}\right)^4 L_n(4|a_1|^2) \exp(-2|\boldsymbol{\alpha}|^2). \quad (5.41)$$

Applying Eqs. (5.34) and (5.35) and integrating over the phase space of one outgoing field, the Wigner function of the field in the j th output channel is derived to be [66]

$$W_{\text{out}}^{(\text{F})}(a_j) = \sum_{k=0}^n \binom{n}{k} |T_{j1}|^{2k} (1 - |T_{j1}|^2)^{n-k} W_k(a_j). \quad (5.42)$$

Obviously, Eq. (5.42) does not only hold for the Wigner function but for any s -parametrized phase-space function, and hence the corresponding density operator reads in the Fock basis as

$$\hat{\rho}_{\text{out } j}^{(\text{F})} = \sum_{k=0}^n \binom{n}{k} |T_{j1}|^{2k} (1 - |T_{j1}|^2)^{n-k} |k\rangle\langle k|. \quad (5.43)$$

Note that the quantum state the outgoing field is prepared in contains only Fock states up to the photon number n of the input Fock state. That is a direct consequence of the action of the compact group $\text{SU}(4)$ which leaves the total number of quanta unchanged. Furthermore, the Fock state with the highest photon number (i.e., the input state) appears in the output state with a weight $|T_{j1}|^{2n}$, that is, the probability of finding the same number of photons after transmission through (reflection at) an absorbing device decreases as $|T_{j1}|^{2n}$. In fact, for a device in the ground state, as it is the case here, there is some classical reasoning explaining this result. One could imagine that each of the n photons “feels” the effect of a transmission (reflection) coefficient smaller than unity, which for n photons amounts to the n th power of the transmission (reflection) coefficient. This reasoning fails when the device is prepared in anything else than the ground state.

Finally, let us assume that the field in one of the two input channels is prepared in a Schrödinger-cat state and the field in the other input channel and the device are left in vacuum, i.e.,

$$|\psi_{\text{in}}\rangle = \frac{1}{\sqrt{N}} (|\gamma\rangle + |-\gamma\rangle) |000\rangle, \quad (5.44)$$

where $|\gamma\rangle$ is a coherent state, and $N = 2[1 + \exp(-2|\gamma|^2)]$ the proper normalization factor. Using Eq. (5.40) with $c_1 = \pm\gamma$, $c_2 = d_1 = d_2 = 0$, it is not difficult to derive the density operator of the field in the j th output channel. The result is

$$\begin{aligned} \hat{\rho}_{\text{out } j}^{(\text{F})} = \frac{1}{N} \Big\{ & |\gamma T_{j1}\rangle\langle\gamma T_{j1}| + |-\gamma T_{j1}\rangle\langle-\gamma T_{j1}| \\ & + (|\gamma T_{j1}\rangle\langle-\gamma T_{j1}| + |-\gamma T_{j1}\rangle\langle\gamma T_{j1}|) \exp[-2|\gamma|^2(1 - |T_{j1}|^2)] \Big\}. \end{aligned} \quad (5.45)$$

¹⁶ $W_{\text{in}}(\boldsymbol{\alpha}) = W_n(a_1)W_0(a_2)W_0(g_1)W_0(g_2)$, where $W_k(a) = (2/\pi)(-1)^k L_k(4|a|^2)e^{-2|a|^2}$ [$L_k(z)$, Laguerre polynomial] is the Wigner function of a k -quanta Fock state (see, e.g., [71]).

Whereas the two peaks decay as $|T_{j1}|^2$, the quantum interference, in contrast, decays exponentially as $|T_{j1}|^2 \exp[-2|\gamma|^2(1 - |T_{j1}|^2)]$. The larger the mean number of photons $\langle \hat{n} \rangle = |\gamma|^2 \tanh |\gamma|^2$ becomes, the faster is the decay. Let us consider, e.g., the transmitted Schrödinger cat ($j=2$). From Eq. (5.13) it follows that the squares of the absolute values of the transmission coefficient $|T_{21}|^2$, the reflection coefficient $|T_{22}|^2$, and the absorption coefficients $|A_{21}|^2$ and $|A_{22}|^2$ are related to each other as $1 - |T_{21}|^2 = |T_{22}|^2 + |A_{21}|^2 + |A_{22}|^2$, so that $\exp[-2|\gamma|^2(1 - |T_{j1}|^2)] = \exp[-2|\gamma|^2|T_{22}|^2] \exp[-2|\gamma|^2(|A_{21}|^2 + |A_{22}|^2)]$. The terms $\exp(-2|\gamma|^2|T_{22}|^2)$ and $\exp[-2|\gamma|^2(|A_{21}|^2 + |A_{22}|^2)]$ then respectively describe the decoherence associated with the losses owing to reflection and absorption.

5.2.2 Entanglement degradation

Quantum information processing such as quantum teleportation and quantum cryptography [72, 73, 74, 75, 76] is essentially based on entanglement, which can be regarded as being the nonclassical contribution to the overall correlation between two parts of a system. In particular, when two fields that are prepared in nonclassical states are superimposed by a four-port device, then the two outgoing fields are prepared in an entangled state in general. Entanglement of a bipartite quantum state $\hat{\rho}$ is commonly quantified by measures which fulfill some basic requirements as non-negativity (being zero only for separable states), invariance under local unitary transformations of the subsystems, and non-increase under arbitrary positive trace-preserving maps [77, 78]. Further, the reduced von Neumann entropy should be recovered for pure states which in addition gives a normalization condition for the measure. So far, the distance $E(\hat{\rho})$ of $\hat{\rho}$ to the set \mathcal{S} of all separable quantum states $\hat{\sigma}$ ¹⁷ which is measured by means of the relative entropy has been proven to be one measure satisfying all the given conditions [77]. Thus,

$$E(\hat{\rho}) = \min_{\hat{\sigma} \in \mathcal{S}} \text{Tr}[\hat{\rho}(\ln \hat{\rho} - \ln \hat{\sigma})]. \quad (5.46)$$

Let us consider the entanglement degradation that is observed when two fields that are prepared in a Bell basis state¹⁸

$$|\Psi^\pm\rangle = \frac{1}{\sqrt{2}} (|01\rangle \pm |10\rangle) \quad (5.47)$$

or

$$|\Phi^\pm\rangle = \frac{1}{\sqrt{2}} (|00\rangle \pm |11\rangle) \quad (5.48)$$

are transmitted through absorbing four-port devices prepared in the ground state.¹⁹ Note that the entanglement of each of the Bell basis states (5.47) and (5.48) is equal

¹⁷A quantum state $\hat{\sigma}$ of a system that consists of two subsystems A and B is called separable if $\hat{\sigma} = \sum_i p_i \hat{\sigma}_i^{(A)} \otimes \hat{\sigma}_i^{(B)}$, where $\hat{\sigma}_i^{(A)}$ and $\hat{\sigma}_i^{(B)}$ represent quantum states of the subsystems, and $\sum_i p_i = 1$.

¹⁸For the entanglement degradation of a two-mode squeezed vacuum, see [79].

¹⁹For an application to photon tunneling through absorbing dielectric barriers, see [80].

to $\ln 2$ (sometimes also named 1 bit). Applying Eqs. (5.32) and (5.33) to the input states $|\psi_{\text{in}}\rangle = |\Psi^\pm\rangle \otimes |0\rangle_{\text{D}}$ and $|\psi_{\text{in}}\rangle = |\Phi^\pm\rangle \otimes |0\rangle_{\text{D}}$ ($|0\rangle_{\text{D}}$, ground state of the two devices), we easily find that the outgoing fields are prepared in the quantum states

$$\begin{aligned} \hat{\rho}_{\text{out}}^{(\text{F})}(|\Psi^\pm\rangle) &= \frac{1}{2} [(2 - |T_1|^2 - |T_2|^2) |00\rangle\langle 00|] \\ &+ \frac{1}{2} (T_2|01\rangle \pm T_1|10\rangle) (T_2^*\langle 01| \pm T_1^*\langle 10|), \end{aligned} \quad (5.49)$$

$$\begin{aligned} \hat{\rho}_{\text{out}}^{(\text{F})}(|\Phi^\pm\rangle) &= \frac{1}{2} [(1 - |T_1|^2) (1 - |T_2|^2) |00\rangle\langle 00| \\ &+ |T_1|^2 (1 - |T_2|^2) |10\rangle\langle 10| + |T_2|^2 (1 - |T_1|^2) |01\rangle\langle 01|] \\ &+ \frac{1}{2} (|00\rangle \pm T_1 T_2 |11\rangle) (\langle 00| \pm (T_1 T_2)^* \langle 11|). \end{aligned} \quad (5.50)$$

Here and in the following the notation $(T_l)_{11} = (T_l)_{22} \equiv R_l$ and $(T_l)_{12} = (T_l)_{21} \equiv T_l$ for the elements of the characteristic transformation matrix \mathbf{T}_l of the l th four-port device is used ($l=1, 2$) [cf. Eqs. (5.14) and (5.15)]. The entanglement contained in the quantum states of the outgoing fields, $\hat{\rho}_{\text{out}}^{(\text{F})}(|\Psi^\pm\rangle)$ and $\hat{\rho}_{\text{out}}^{(\text{F})}(|\Phi^\pm\rangle)$, can be estimated by using the convexity property of the relative entropy.²⁰ In Eqs. (5.49) and (5.50) the output state is written as a sum of separable states and a single pure state. Since separable states have zero entanglement by definition, the entanglement of the whole state is bounded from above by the reduced von Neumann entropy of the respective pure states. If we assume equal transmission coefficients of the two devices ($T_1 = T_2 = T$), the bounds are given according to [70]

$$E[\hat{\rho}_{\text{out}}^{(\text{F})}(|\Psi^\pm\rangle)] \leq |T|^2 \ln 2 \quad (5.51)$$

and

$$E[\hat{\rho}_{\text{out}}^{(\text{F})}(|\Phi^\pm\rangle)] \leq \frac{1}{2} [(1 + |T|^4) \ln(1 + |T|^4) - |T|^4 \ln |T|^4]. \quad (5.52)$$

Suppose the two fields are transmitted through (equal) optical fibres with perfect input coupling ($R=0$), so that the transmission coefficient T may be given by

$$T = e^{i n \omega l / c} = e^{i n_R \omega l / c} e^{-l/L}, \quad (5.53)$$

with l and $L = c/(n_l \omega)$ being respectively the propagation length through the fibres and the absorption length of the fibres. Substituting of this expression into the inequalities (5.51) and (5.52) yields the dependence on l of the bounds of entanglement. The exact dependence on l of the entanglement degradation calculated by applying Eq. (5.46) is shown in Fig. 3. One observes that the states $|\Phi^\pm\rangle$ decay faster than the states $|\Psi^\pm\rangle$. Since the device has been left in the ground state, we can again use some classical reasoning to explain this behaviour. When the two fields are initially

²⁰ $E(\sum_i p_i \hat{\rho}_i) \leq \sum_i p_i E(\hat{\rho}_i)$, where $\sum_i p_i = 1$ [81]. The partition into separable states and a single pure state is not unique. It has been proven, however, that for a pair of spin- $\frac{1}{2}$ parties there exists a unique (sometimes called optimal) decomposition such that the weight of the separable state is maximized [82]. The reduced von Neumann entropy of the extracted pure state with corresponding minimal weight is then exactly the amount of entanglement, hence the inequality reduces to an equality.

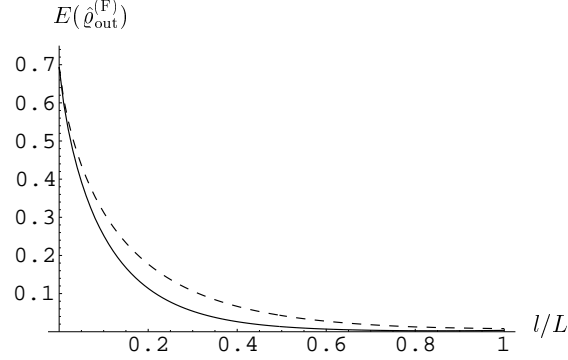


Figure 3: Entanglement degradation of Bell basis states $|\Phi^\pm\rangle$ (full curve) and $|\Psi^\pm\rangle$ (dashed curve) after transmission through absorbing fibres as a function of the fibre length.

prepared in a state $|\Psi^\pm\rangle$, then only a single photon (i.e., either the photon from the first field or the photon from the second one) is effectively subject to absorption. By contrast, the two photons get effected simultaneously, if the initial state is a state $|\Phi^\pm\rangle$.

6 Spontaneous decay

Spontaneous emission is a prime example of the action of ground-state fluctuations on physically measurable processes. Einstein [83] already pointed out that, in order to obtain the Planck radiation law, a process as spontaneous emission must necessarily be included in the theory of atomic decay. Later on, the radiation properties of an excited atom located in free space have been a subject of many studies. In particular, the rate of spontaneous emission of an excited (two-level) atom in free space is given by (the famous Einstein A -coefficient)

$$\Gamma_0 = \frac{\omega_A^3 d^2}{3\pi\hbar\epsilon_0 c^3}, \quad (6.1)$$

where d is the absolute value of the matrix element $\mathbf{d} = \langle l | \hat{\mathbf{d}}_A | u \rangle$ of the atomic dipole operator $\hat{\mathbf{d}}_A$ [Eq. (6.6)] between the upper state $|u\rangle$ and the lower state $|l\rangle$, and ω_A is the corresponding atomic transition frequency. When the atom is surrounded by (dielectric) matter, then the ground state felt by the radiating atom is changed and thus the rate formula (6.1) must be corrected²¹ [17, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98].

²¹In the strong-coupling regime the decay becomes non-exponential and cannot be described by a rate (Sec. 6.4).

6.1 Time dependence of the atom–field system

6.1.1 Hamiltonian

Let us assume that the surrounding matter can be regarded as being a dielectric of given complex permittivity. Applying the minimal-coupling scheme (Sec. 4.1), we may decompose the Hamiltonian [Eq. (4.1)] of the coupled system consisting of an atom and the medium-assisted electromagnetic field as

$$\hat{H} = \hat{H}_A + \hat{H}_M + \hat{H}_{\text{int}}, \quad (6.2)$$

where

$$\hat{H}_A = \sum_{\alpha} \frac{\hat{\mathbf{p}}_{\alpha}^2}{2m_{\alpha}} + \frac{1}{2} \int d^3\mathbf{r} \hat{\rho}_A(\mathbf{r}) \hat{\varphi}_A(\mathbf{r}) \quad (6.3)$$

is the Hamiltonian of the atom,

$$\hat{H}_M = \int d^3\mathbf{r} \int_0^{\infty} d\omega \hbar\omega \hat{\mathbf{f}}^{\dagger}(\mathbf{r}, \omega) \hat{\mathbf{f}}(\mathbf{r}, \omega) \quad (6.4)$$

is the Hamiltonian of the electromagnetic field and the dielectric matter, and

$$\hat{H}_{\text{int}} = - \sum_{\alpha} \frac{q_{\alpha}}{m_{\alpha}} \hat{\mathbf{p}}_{\alpha} \hat{\mathbf{A}}(\mathbf{r}_{\alpha}) + \int d^3\mathbf{r} \hat{\rho}_A(\mathbf{r}) \hat{\varphi}_M(\mathbf{r}) \quad (6.5)$$

is the interaction energy. Here we have omitted the $\hat{\mathbf{A}}^2$ term. In the electric-dipole approximation, the first term on the right-hand side of Eq. (6.5) simplifies to

$$- \sum_{\alpha} \frac{q_{\alpha}}{m_{\alpha}} \hat{\mathbf{p}}_{\alpha} \hat{\mathbf{A}}(\mathbf{r}_{\alpha}) = - \frac{1}{i\hbar} [\hat{\mathbf{d}}_A, \hat{H}_A] \hat{\mathbf{A}}(\mathbf{r}_A), \quad (6.6)$$

where

$$\hat{\mathbf{d}}_A = \sum_{\alpha} q_{\alpha} \hat{\mathbf{r}}_{\alpha} \quad (6.7)$$

is the atomic dipole operator. Restricting our attention to a two-level system, the atomic Hamiltonian \hat{H}_A reduces to

$$\hat{H}_A = \hbar\omega_u |u\rangle\langle u| + \hbar\omega_l |l\rangle\langle l| = \frac{1}{2} \hbar\omega_A \hat{\sigma}_z + \text{const.}, \quad (6.8)$$

where $\hat{\sigma}_z = |u\rangle\langle u| - |l\rangle\langle l|$. Thus we may further simplify Eq. (6.6) to obtain, on applying the rotating wave approximation and recalling Eqs. (3.46) and (3.47),

$$- \sum_{\alpha} \frac{q_{\alpha}}{m_{\alpha}} \hat{\mathbf{p}}_{\alpha} \hat{\mathbf{A}}(\mathbf{r}_{\alpha}) = - \hat{\sigma}^{\dagger} \hat{\mathbf{E}}_M^{\perp(+)}(\mathbf{r}_A) \mathbf{d} + \text{H.c.} \quad (6.9)$$

(\mathbf{d} real), with $\hat{\sigma}$ being defined by $\hat{\sigma} = |l\rangle\langle u|$.

For a neutral atom, the atomic polarization defined by Eq. (4.14) reduces in the dipole approximation to

$$\hat{\mathbf{P}}_A(\mathbf{r}) = \hat{\mathbf{d}}_A \delta(\mathbf{r} - \mathbf{r}_A), \quad (6.10)$$

and thus application of Eq. (4.24) yields ($\hat{\mathbf{E}}_{\text{M}}^{\parallel} = -\hat{\mathbf{P}}_{\text{M}}^{\parallel}/\varepsilon_0$)

$$\int d^3\mathbf{r} \hat{\rho}_{\text{A}}(\mathbf{r}) \hat{\varphi}_{\text{M}}(\mathbf{r}) = -\hat{\mathbf{d}}_{\text{A}} \hat{\mathbf{E}}_{\text{M}}^{\parallel}(\mathbf{r}_{\text{A}}), \quad (6.11)$$

which for a two-level atom in the rotating wave approximation reads

$$\int d^3\mathbf{r} \hat{\rho}_{\text{A}}(\mathbf{r}) \hat{\varphi}_{\text{M}}(\mathbf{r}) = -\hat{\sigma}^{\dagger} \hat{\mathbf{E}}_{\text{M}}^{\parallel(+)}(\mathbf{r}_{\text{A}}) \mathbf{d} + \text{H.c.} \quad (6.12)$$

Eqs. (6.9) and (6.12) yield

$$\hat{H}_{\text{int}} = -\hat{\sigma}^{\dagger} \hat{\mathbf{E}}_{\text{M}}^{(+)}(\mathbf{r}_{\text{A}}) \mathbf{d} + \text{H.c.} \quad (6.13)$$

6.1.2 Heisenberg picture

From the Hamiltonian given in Eq. (6.2) together with Eqs. (6.4), (6.8), and (6.13), the Heisenberg equations of motion read [95]

$$\dot{\hat{\sigma}}_z = \frac{2i}{\hbar} \hat{\sigma}^{\dagger} \hat{\mathbf{E}}_{\text{M}}^{(+)}(\mathbf{r}_{\text{A}}) \mathbf{d} + \text{H.c.}, \quad (6.14)$$

$$\dot{\hat{\sigma}}^{\dagger} = i\omega_{\text{A}} \hat{\sigma}^{\dagger} + \frac{i}{\hbar} \hat{\mathbf{E}}_{\text{M}}^{(-)}(\mathbf{r}_{\text{A}}) \mathbf{d} \hat{\sigma}_z, \quad (6.15)$$

$$\dot{\hat{\mathbf{f}}}(\mathbf{r}, \omega) = -i\omega \hat{\mathbf{f}}(\mathbf{r}, \omega) + \frac{\omega^2}{c^2} \sqrt{\frac{\varepsilon_{\text{I}}(\mathbf{r}, \omega)}{\hbar\pi\varepsilon_0}} \mathbf{G}^*(\mathbf{r}, \mathbf{r}_{\text{A}}, \omega) \mathbf{d} \hat{\sigma}. \quad (6.16)$$

Substituting in the expression for the electric field strength [Eq. (3.38) together with Eq. (3.35)] for $\hat{\mathbf{f}}(\mathbf{r}, \omega)$ the formal solution of Eq. (6.16), we derive, on using the relation (3.28),

$$\begin{aligned} \hat{\mathbf{E}}_{\text{M}}^{(+)}(\mathbf{r}, t) &= \hat{\mathbf{E}}_{\text{M free}}^{(+)}(\mathbf{r}, t) \\ &+ \frac{i}{\pi\varepsilon_0} \int_0^\infty d\omega \frac{\omega^2}{c^2} \text{Im} \mathbf{G}(\mathbf{r}, \mathbf{r}_{\text{A}}, \omega) \mathbf{d} \int_{t'}^t d\tau e^{-i\omega(t-\tau)} \hat{\sigma}(\tau). \end{aligned} \quad (6.17)$$

Substitution of this expression into Eqs. (6.14) and (6.15) then yields a system of integro-differential equations for the atomic quantities.

In the Markov approximation the integro-differential equations reduce to Langevin-type differential equations. It is assumed that [after performing the ω -integration in Eq. (6.17)] the time integral effectively runs over a small correlation time interval τ_c . As long as we require that $t - t' \gg \tau_c$, we may extend the lower limit of the τ -integral to minus infinity with little error. Further we require that τ_c be small on a time scale on which the atomic system is changed owing to the coupling to the (medium-assisted) electromagnetic field. In this case, in the τ -integral in Eq. (6.17) the slowly varying atomic quantity $\hat{\sigma}(\tau)e^{i\omega_{\text{A}}\tau}$ can be taken at time t and put in front of the integral, thus

$$\hat{\mathbf{E}}_{\text{M}}^{(+)}(\mathbf{r}, t) = \hat{\mathbf{E}}_{\text{M free}}^{(+)}(\mathbf{r}, t) + \hat{\sigma}(t) \frac{i}{\pi\varepsilon_0} \int_0^\infty d\omega \frac{\omega^2}{c^2} \text{Im} \mathbf{G}(\mathbf{r}, \mathbf{r}_{\text{A}}, \omega) \mathbf{d} \zeta(\omega_{\text{A}} - \omega) \quad (6.18)$$

$[\zeta(x) = \pi\delta(x) + i\mathcal{P}x^{-1}]$. Substitution of this expression into Eqs. (6.14) and (6.15) yields

$$\dot{\hat{\sigma}}_z = -\Gamma(1 + \hat{\sigma}_z) + \left[\frac{2i}{\hbar} \hat{\sigma}^\dagger \hat{\mathbf{E}}_{\text{M free}}^{(+)}(\mathbf{r}_A) \mathbf{d} + \text{H.c.} \right], \quad (6.19)$$

$$\dot{\hat{\sigma}}^\dagger = [i(\omega_A - \delta\omega) - \tfrac{1}{2}\Gamma] \hat{\sigma}^\dagger + \frac{i}{\hbar} \hat{\mathbf{E}}_{\text{M free}}^{(-)}(\mathbf{r}_A) \mathbf{d} \hat{\sigma}_z, \quad (6.20)$$

where

$$\Gamma = \frac{2\omega_A^2 d_i d_j}{\hbar \epsilon_0 c^2} \text{Im } G_{ij}(\mathbf{r}_A, \mathbf{r}_A, \omega_A) \quad (6.21)$$

is the rate of spontaneous decay of the upper state and

$$\delta\omega = \frac{d_i d_j}{\hbar \pi \epsilon_0} \mathcal{P} \int_0^\infty d\omega \frac{\omega^2}{c^2} \frac{\text{Im } G_{ij}(\mathbf{r}_A, \mathbf{r}_A, \omega)}{\omega - \omega_A} \quad (6.22)$$

is the contribution to the Lamb shift.²² Note that from Eq. (3.44) it follows that Eq. (6.21) can be given in the equivalent form of

$$\Gamma = \frac{2\pi}{\hbar^2} d_i d_j \int_0^\infty d\omega \langle 0 | \hat{\underline{E}}_i(\mathbf{r}_A, \omega) \hat{\underline{E}}_j^\dagger(\mathbf{r}_A, \omega_A) | 0 \rangle, \quad (6.23)$$

which exactly corresponds to Fermi's Golden Rule (see, e.g., [100]).

6.1.3 Schrödinger picture

The above given equations of motion do not only apply to the spontaneous emission but are also suitable for the study of the evolution of a two-level atom driven by an external medium-assisted electromagnetic field. If the atom is initially prepared in the upper state and there is no driving field, then the use of the wave equation

$$i\hbar \frac{d}{dt} |\Psi\rangle = \hat{H} |\Psi\rangle \quad (6.24)$$

may be more appropriate for the study of the motion of the coupled atom-field system [101]. According to the Hamiltonian in Eq. (6.2) together with Eqs. (6.4), (6.8), and (6.13), the state vector $|\Psi(t)\rangle$ can be expanded as

$$\begin{aligned} |\Psi(t)\rangle &= C_u(t) e^{-i\omega_u t} |u\rangle \otimes |0\rangle \\ &+ \int d^3\mathbf{r} \int_0^\infty d\omega C_{li}(\mathbf{r}, \omega, t) e^{-i(\omega+\omega_l)t} |l\rangle \otimes |i, \mathbf{r}, \omega\rangle, \end{aligned} \quad (6.25)$$

where $|0\rangle$ is the vacuum state of the fundamental fields $\hat{f}_i(\mathbf{r}, \omega)$, and $|i, \mathbf{r}, \omega\rangle$ is the state, where one of them is excited in a single-quantum Fock state. It is not difficult to prove that the probability amplitudes C_u and C_{li} satisfy the differential equations

$$\begin{aligned} \dot{C}_u(t) &= -\frac{d_j}{\sqrt{\pi\epsilon_0\hbar}} \int_0^\infty d\omega \frac{\omega^2}{c^2} \int d^3\mathbf{r} \left[\sqrt{\epsilon_1(\mathbf{r}, \omega)} \right. \\ &\quad \left. \times G_{ji}(\mathbf{r}_A, \mathbf{r}, \omega) C_{li}(\mathbf{r}, \omega, t) e^{-i(\omega-\omega_A)t} \right], \end{aligned} \quad (6.26)$$

²²For the the overall (vacuum) Lamb shift, see, e.g., [99].

$$\dot{C}_{li}(\mathbf{r}, \omega, t) = \frac{d_j}{\sqrt{\pi\epsilon_0\hbar}} \frac{\omega^2}{c^2} \sqrt{\epsilon_I(\mathbf{r}, \omega)} G_{ji}^*(\mathbf{r}_A, \mathbf{r}, \omega) C_u(t) e^{i(\omega-\omega_A)t}. \quad (6.27)$$

We now substitute the result of formal integration of Eq. (6.27) [$C_{li}(\mathbf{r}, \omega, 0) = 0$] into Eq. (6.26). Making use of the relationship (3.28), we obtain the integro-differential equation

$$\dot{C}_u(t) = \int_0^t dt' K(t-t') C_u(t'), \quad (6.28)$$

with the kernel function

$$K(t-t') = -\frac{d_i d_j}{\hbar\pi\epsilon_0} \int_0^\infty d\omega \frac{\omega^2}{c^2} \text{Im} G_{ij}(\mathbf{r}_A, \mathbf{r}_A, \omega) e^{-i(\omega-\omega_A)(t-t')}. \quad (6.29)$$

Taking the time integral of both sides of Eq. (6.28), we easily derive, on changing the order of integrations on the right-hand side,

$$C_u(t) = \int_0^t dt' \bar{K}(t-t') C_u(t') + 1 \quad (6.30)$$

[$C_u(0) = 1$], where

$$\bar{K}(t-t') = \frac{d_i d_j}{\hbar\pi\epsilon_0} \int_0^\infty d\omega \frac{\omega^2}{c^2} \frac{\text{Im} G_{ij}(\mathbf{r}_A, \mathbf{r}_A, \omega)}{i(\omega - \omega_A)} \left[e^{-i(\omega-\omega_A)(t-t')} - 1 \right]. \quad (6.31)$$

Note that in the Markov approximation the kernel in Eq. (6.31) simply becomes

$$K(t-t') = -\frac{1}{2}\Gamma + i\delta\omega, \quad (6.32)$$

where Γ and $\delta\omega$ are respectively given by Eqs. (6.21) and (6.22).

The equation (6.30) is a well-known Volterra integral equation of the second kind. It is worth noting that the integro-differential equation (6.28) and the equivalent integral equation (6.30) apply to the spontaneous decay of an atom in the presence of an arbitrary configuration of dispersing and absorbing dielectric bodies. All the matter parameters that are relevant for the atomic evolution are contained, via the Green tensor, in the kernel functions (6.29) and (6.31). In particular when absorption is disregarded and the permittivity is regarded as being a real frequency-independent quantity (which of course can change with space), then the formalism yields the results of standard mode decomposition (see, e.g. [102, 103, 104]).

6.2 Atom near a dielectric surface

As a first example, let us consider the spontaneous decay of an excited atom near an absorbing planar dielectric surface. To be more specific, the distance z to the surface of the atom is assumed to be small compared to the atomic transition wavelength. For real permittivity, this configuration has been studied extensively in connection with Casimir and van der Waals forces (see, e.g., [85, 88] and references cited therein).

Further, it can be regarded as being the basic configuration in scanning near-field optical microscopy (see, e.g., [105]) and related applications.

In order to calculate the decay rate Γ , Eq. (6.21), the imaginary part of the Green tensor of two (infinite) half-spaces with a common interface [106, 107] is required. When the atom's half-space is the vacuum, then the Green tensor in this half-space can be decomposed into the vacuum Green tensor and a reflection term, i.e.,

$$G_{kl}(\mathbf{r}, \mathbf{r}', \omega) = G_{kl}^V(\mathbf{r}, \mathbf{r}', \omega) + R_{kl}(\mathbf{r}, \mathbf{r}', \omega), \quad (6.33)$$

where the vacuum Green tensor $G_{kl}^V(\mathbf{r}, \mathbf{r}', \omega)$ is obtained from Eq. (A.34) for $q = \omega/c$. From Eq. (A.36) it is seen that $\text{Im } G_{kl}^{V\parallel}(\mathbf{r}, \mathbf{r}', \omega) = 0$, and thus we obtain, on applying Eq. (A.38),

$$\text{Im } G_{kl}^V(\mathbf{r}, \mathbf{r}, \omega) = \frac{\omega}{6\pi c} \delta_{kl}. \quad (6.34)$$

Combining Eqs. (6.21), (6.33), and (6.34), we obtain

$$\Gamma = \Gamma_0 + \frac{2\omega_A^2 d_k d_l}{\hbar \varepsilon_0 c^2} \text{Im } R_{kl}(\mathbf{r}_A, \mathbf{r}_A, \omega_A), \quad (6.35)$$

where Γ_0 is given by Eq. (6.1)

In the coincidence limit, the reflection term $R_{kl}(\mathbf{r}, \mathbf{r}, \omega)$ can be given in the form [39, 108]

$$\begin{aligned} R_{xx}(z, z, \omega) &= R_{yy}(z, z, \omega) \\ &= -\frac{i}{8\pi q^2} \int_0^\infty dk k \beta e^{2i\beta z} r^p(k) + \frac{i}{8\pi} \int_0^\infty dk \frac{k e^{2i\beta z}}{\beta} r^s(k), \end{aligned} \quad (6.36)$$

$$R_{zz}(z, z, \omega) = \frac{i}{4\pi q^2} \int_0^\infty dk k^3 \frac{e^{2i\beta z}}{\beta} r^p(k), \quad (6.37)$$

where $r^p(k)$ and $r^s(k)$ are the usual Fresnel reflection coefficients for p - and s -polarized waves ($q = \omega/c$, $\beta = \sqrt{q^2 - k^2}$; the origin of the co-ordinates is on the interface). When the distance z of the atom from the surface is small compared to the wavelength, $qz \ll 1$, then the integrals in Eqs.(6.36) and (6.37) can be evaluated asymptotically to obtain

$$R_{zz} = \frac{1}{16\pi q^2 z^3} \frac{n^2 - 1}{n^2 + 1} + \frac{1}{8\pi z} \frac{(n - 1)^2}{n(n + 1)} + \frac{iq}{12\pi} \frac{(n - 1)(2n - 1)}{n(n + 1)} + O(z), \quad (6.38)$$

$$R_{xx} = R_{yy} = \frac{1}{2} R_{zz} - \frac{1}{16\pi z} \frac{n^2 - 1}{n^2 + 1} - \frac{iq}{3\pi} \frac{n - 1}{n + 1} + O(z). \quad (6.39)$$

$[\varepsilon = \varepsilon(\omega), n = \sqrt{\varepsilon(\omega)}]$. Inserting Eqs. (6.38) and (6.39) into Eq. (6.35) yields ($\omega = \omega_A$) [109]

$$\Gamma = \Gamma_0 \frac{3}{8} \left(1 + \frac{d_z^2}{d^2} \right) \left(\frac{c}{\omega_A z} \right)^3 \frac{\varepsilon_I(\omega_A)}{|\varepsilon(\omega_A) + 1|^2} + O(z^{-1}). \quad (6.40)$$

It is worth noting that the leading term $\sim z^{-3}$ in Eq. (6.40) exactly agrees with the result of the involved microscopic approach in [91]. It typically occurs for absorbing media and reflects the possibility of nonradiative decay, if the excited atom is sufficiently near the medium.

6.3 Real-cavity model of spontaneous decay in a dielectric medium

Let us consider the situation when an excited atom is placed inside an empty microsphere (embedded in an otherwise homogeneous dielectric) with a radius much smaller than the wavelength of the atomic transition. This might serve as a model for spontaneous decay of a (guest) atom in an absorbing dielectric.²³ From simple arguments based on the change of the mode density it is suggested that the spontaneous emission rate inside a nonabsorbing dielectric should be modified according to $\Gamma = n\Gamma_0$, where n is the (real) refractive index of the medium and Γ_0 is given by Eq. (6.1) [110, 111, 112, 113]. Here it is assumed that the local field the atom interacts with is exactly the same as the electromagnetic field in the continuous medium. In reality, the atom is located in a small region of free space, and the local field is different from the field in the continuous medium which leads to the introduction of the so-called local-field correction factor ξ , thus giving $\Gamma = n\xi\Gamma_0$. Different models have been used to compute ξ . Here we want to concentrate on the (Glauber-Lewenstein) real-cavity model, which for real n leads to [10]

$$\xi = \left(\frac{3n^2}{2n^2 + 1} \right)^2. \quad (6.41)$$

Experiments suggest that this model is a good candidate for describing the decay of substitutional guest atoms different from the constituents of the dielectric [114, 115, 116].

Now let us turn to the question what will appear for absorbing media. According to Eq. (6.21), the rate of spontaneous decay is proportional to the imaginary part of the Green tensor in the coincidence limit (i.e., the two spatial arguments are to be taken at the position of the atom). In the real-cavity model the Green tensor that enters the rate formula is the Green tensor for the system disturbed by a small free-space inhomogeneity. Obviously, it can again be given in a form like in Eq. (6.33), where for a sphere (with the centre as origin of co-ordinates) the reflection term $\mathbf{R}(\mathbf{r}, \mathbf{r}', \omega)$ reads [117]

$$\begin{aligned} \mathbf{R}(\mathbf{r}, \mathbf{r}', \omega) = & \frac{i\omega}{4\pi c} \sum_{e,o} \sum_{n=1}^{\infty} \sum_{m=0}^n \left\{ \frac{2n+1}{n(n+1)} \frac{(n-m)!}{(n+m)!} (2 - \delta_{0m}) \right. \\ & \times \left[C_n^M(\omega) \mathbf{M}_{onm}^e\left(\mathbf{r}, \frac{\omega}{c}\right) \otimes \mathbf{M}_{onm}^e\left(\mathbf{r}', \frac{\omega}{c}\right) \right. \\ & \left. \left. + C_n^N(\omega) \mathbf{N}_{onm}^e\left(\mathbf{r}, \frac{\omega}{c}\right) \otimes \mathbf{N}_{onm}^e\left(\mathbf{r}', \frac{\omega}{c}\right) \right] \right\}, \end{aligned} \quad (6.42)$$

where $\mathbf{M}_{onm}^e(\mathbf{r}, k)$ and $\mathbf{N}_{onm}^e(\mathbf{r}, k)$ are the (even and odd) vector Debye potentials defined by

$$\mathbf{M}_{onm}^e(\mathbf{r}, k) = \nabla \times \left[\psi_{onm}^e(\mathbf{r}, k) \mathbf{r} \right], \quad (6.43)$$

²³Such a model applies as long as the atom cannot “resolve” the atomic structure of the surrounding matter.

$$\mathbf{N}_{\circ nm}^e(\mathbf{r}, k) = \frac{1}{k} \nabla \times \nabla \times [\psi_{\circ nm}^e(\mathbf{r}, k) \mathbf{r}], \quad (6.44)$$

with

$$\psi_{\circ nm}^e(\mathbf{r}, k) = j_n(kr) P_n^m(\cos \Theta) \begin{pmatrix} \cos \\ \sin \end{pmatrix} (m\phi) \quad (6.45)$$

$[j_n(kr)]$, spherical Bessel function of the first kind; $P_n^m(\cos \Theta)$, associated Legendre polynomial; for the rather lengthy expressions of the generalized reflection coefficients $C_n^{M(N)}$, see [117].

In particular in the coincidence limit $\mathbf{r} \rightarrow \mathbf{r}' \rightarrow 0$ only the TM-wave vector Debye potentials $\mathbf{N}_{\circ 10}^e(\mathbf{r}, k)$ and $\mathbf{N}_{\circ 11}^e(\mathbf{r}, k)$ contribute to $R_{kl}(\mathbf{r}, \mathbf{r}', \omega)$ and we find that

$$R_{kl}(\mathbf{r}, \mathbf{r}, \omega)|_{\mathbf{r}=0} = \frac{i\omega}{6\pi c} C_1^N(\omega) \delta_{kl}, \quad (6.46)$$

where the generalized reflection coefficient $C_1^N(\omega)$ reads

$$C_1^N(\omega) = \frac{[i + \varrho(n+1) - i\varrho^2 n - \varrho^3 n^2/(n+1)] e^{i\varrho}}{\sin \varrho - \varrho(\cos \varrho + in \sin \varrho) + i\varrho^2 n \cos \varrho - \varrho^3(\cos \varrho - in \sin \varrho)n^2/(n^2-1)} \quad (6.47)$$

$[n = \sqrt{\varepsilon(\omega)}$ and $\varrho = R\omega/c$, with R being the cavity radius]. Hence, when the atom is located at the centre of the sphere, we may insert Eq. (6.46) into Eq. (6.35) to obtain (for $\omega = \omega_A$) the decay rate in the form of²⁴ [95]

$$\Gamma = \Gamma_0 [1 + \text{Re } C_1^N(\omega_A)] . \quad (6.48)$$

As long as the surrounding medium can be treated as a continuum this result is exact and valid for all admissible cavity sizes, without restriction to transition frequencies far from medium resonances. If the cavity radius is small compared to the wavelength of the atomic transition (but large enough for the continuum approach), we may expand the generalized reflection coefficient $C_1^N(\omega_A)$, Eq. (6.47), in powers of $\varrho = R\omega_A/c$ to obtain

$$\begin{aligned} \Gamma = \Gamma_0 & \left\{ \frac{9\varepsilon_I(\omega_A)}{|2\varepsilon(\omega_A) + 1|^2} \left(\frac{c}{\omega_A R} \right)^3 \right. \\ & + \frac{9\varepsilon_I(\omega_A) [28|\varepsilon(\omega_A)|^2 + 16\varepsilon_R(\omega_A) + 1]}{5|2\varepsilon(\omega_A) + 1|^4} \left(\frac{c}{\omega_A R} \right) \\ & + \frac{9n_R(\omega_A)}{|2\varepsilon(\omega_A) + 1|^4} [4|\varepsilon(\omega_A)|^4 + 4\varepsilon_R(\omega_A)|\varepsilon(\omega_A)|^2 + \varepsilon_R^2(\omega_A) - \varepsilon_I^2(\omega_A)] \\ & \left. - \frac{9n_I(\omega_A)\varepsilon_I(\omega_A)}{|2\varepsilon(\omega_A) + 1|^4} [4|\varepsilon(\omega_A)|^2 + 2\varepsilon_R(\omega_A)] \right\} + O\left(\frac{R\omega_A}{c}\right) . \quad (6.49) \end{aligned}$$

²⁴Note that when in the coincidence limit (at the position of the atom) the Green tensor has the form of Eq. (6.33), then the rate of the spontaneous decay has the form of Eq. (6.35). Since the vacuum Green tensor $G_{kl}^V(\mathbf{r}, \mathbf{r}', \omega)$ has no longitudinal imaginary part and the scattering component $R_{kl}(\mathbf{r}, \mathbf{r}', \omega)$ is purely transverse, the longitudinal electromagnetic field does not contribute to the decay rate.

For $\varepsilon_I(\omega_A) = 0$, i.e, when absorption is fully disregarded, in Eq. (6.49) only the third term survives and reproduces exactly the Glauber-Lewenstein local-field correction factor (6.41). For an absorbing medium terms proportional to R^{-3} and R^{-1} are observed, which can give rise to a strong dependence of the decay rate on the cavity radius. In particular, the leading term proportional to R^{-3} can be regarded as corresponding to nonradiative decay via dipole-dipole energy transfer from the atom to the medium.

Examples of the dependence of rate of spontaneous decay on the atomic transition frequency are plotted in Fig. 4 for a model permittivity of Lorentz type

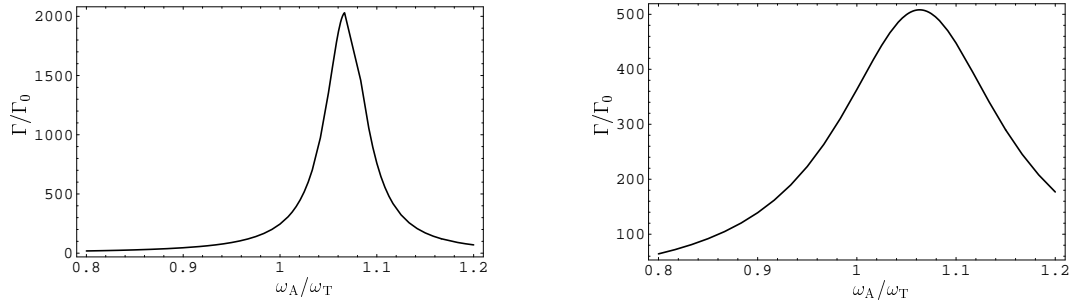


Figure 4: The spontaneous decay rate Γ , Eq. (6.48), for the real-cavity model of an atom in an absorbing dielectric is shown as a function of the atomic transition frequency ω_A near a medium resonance for the model permittivity (6.50) ($\omega_P = 0.46 \omega_T$, $\gamma = 0.05 \omega_T$ [left figure] and $\gamma = 0.2 \omega_T$ [right figure]) and $R = 0.02 \lambda_A$.

$$\varepsilon(\omega) = 1 + \frac{\omega_P^2}{\omega_T^2 - \omega^2 - i\gamma\omega}. \quad (6.50)$$

It shows a band gap between the (transverse) resonance frequency ω_T and the longitudinal frequency $\omega_L = \sqrt{\omega_T^2 + \omega_P^2}$. One observes, that precisely in the gap region the spontaneous decay strongly increases due to non-radiative decay channels which become important in that frequency range.

6.4 Cavity QED

If the radius of the microcavity is not small compared with the wavelength of the atomic transition, the cavity can act as a resonator, and it is well known that the spontaneous decay of an excited atom can be strongly modified when it is placed in a microresonator (see, e.g., [118, 119]). There are typically two qualitatively different regimes: the weak coupling regime and the strong-coupling regime. In the weak coupling regime the Markov approximation applies and a monotonous exponential decay is observed, the decay rate being enhanced or reduced compared to the free-space value depending on whether the atomic transition frequency fits a cavity resonance or not. The strong-coupling regime, in contrast, is characterized by reversible Rabi

oscillations where the energy of the initially excited atom is periodically exchanged between the atom and the radiation field. This usually requires that the emission is in resonance with a high-quality cavity mode. Recent progress in constructing certain types of microresonators such as microspheres has rendered it possible to approach the ultimate quality level determined by intrinsic material losses [120].

Let us consider the spontaneous decay of an excited atom placed inside a spherical three-layer structure (Fig. 5). The outer layer ($r > R_1$) and the inner layer ($0 \leq r < R_2$) are vacuum, whereas the middle layer ($R_2 \leq r \leq R_1$), which plays the role of the resonator wall, is a dispersive and absorbing dielectric. In particular when a Lorentz-type dielectric is assumed whose permittivity is of the form (6.50), the wall would be perfectly reflecting in the band-gap zone, i.e., $\omega_T < \omega_A < \omega_L$, provided that absorption could be disregarded ($\gamma \rightarrow 0$). The Green tensor $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ for a spherical three-layer geometry can be found in [117].

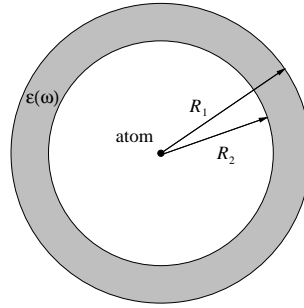


Figure 5: Scheme of the spherical microresonator.

6.4.1 Weak coupling

For \mathbf{r} and \mathbf{r}' inside the inner (vacuum) sphere, the Green tensor looks like the one for the two-layered spherical geometry considered in Sec. 6.3. It has again the form given in Eq. (6.33) together with Eqs. (6.42) – (6.45), but with different reflection coefficients $C_n^{M(N)}$ (see [117]). Hence, when the atom is located at the centre of the inner sphere, then the rate of spontaneous decay can again be given in the form of Eq. (6.48). Obviously, for a sufficiently thick cavity wall, $\exp[-in_I(\omega_A)(R_1 - R_2)\omega/c] \ll 1$, the generalized reflection coefficient C_1^N in Eq. (6.48) reduces to that one for the two-layered configuration [Eq. (6.47) with $R = R_2$], and thus for a true microresonator, $R_2\omega_A/c \gg 1$, the decay rate becomes [101]

$$\begin{aligned} \Gamma &\simeq \Gamma_0 \operatorname{Re} \left[\frac{n(\omega_A) - i \tan(R_2\omega_A/c)}{1 - in(\omega_A) \tan(R_2\omega_A/c)} \right] \\ &= \Gamma_0 \frac{n_R(\omega_A)[1 + \tan^2(R_2\omega_A/c)]}{[1 + n_I(\omega_A) \tan(R_2\omega_A/c)]^2 + n_R^2(\omega_A) \tan^2(R_2\omega_A/c)}. \end{aligned} \quad (6.51)$$

From Fig. 6 it is seen that [for the model permittivity (6.50)] the rate of spontaneous

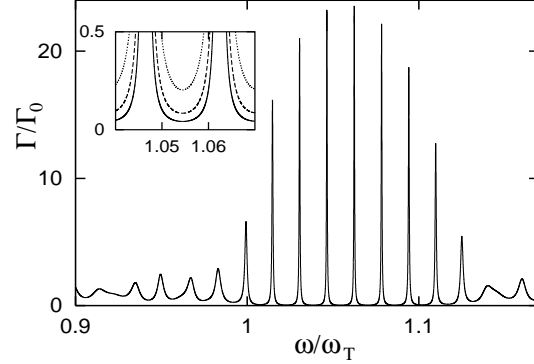


Figure 6: The spontaneous decay rate Γ/Γ_0 , Eq. (6.48), of an atom in the microresonator in Fig. 5 is shown for $R_2 = 30\lambda_T$, $R_1 - R_2 = \lambda_T$, $\omega_P = 0.5\omega_T$, and $\gamma = 10^{-2}\omega_T$. The curves in the inset correspond to $\gamma = 10^{-2}\omega_T$ (solid line), $\gamma = 2 \times 10^{-2}\omega_T$ (dashed line), and $\gamma = 5 \times 10^{-2}\omega_T$ (dotted line).

decay sensitively depends on the transition frequency. Narrow-band enhancement of spontaneous decay ($\Gamma/\Gamma_0 > 1$) alternates with broadband inhibition ($\Gamma/\Gamma_0 < 1$). The frequencies where the maxima of enhancement are observed correspond to the resonance frequencies of the cavity. Within the band gap the heights and widths of the frequency intervals in which spontaneous decay is feasible are essentially determined by the material losses. Outside the band-gap zone the change of the decay rate is less pronounced, because of the relatively large input–output coupling, the (small) material losses being of secondary importance.

6.4.2 Strong coupling

The strength of the coupling between the atom and the electromagnetic field increases when the frequency of the atomic transition frequency approaches a cavity resonance frequency. In order to gain insight into the solution of the integral equation (6.30) for the strong-coupling regime, let us consider the limiting case of only a single cavity resonance (frequency ω_C) being involved in the atom–field interaction. In this case we may approximate, on using Eqs. (6.21) and (6.48), the kernel function (6.31) by

$$\begin{aligned} \bar{K}(t-t') &\simeq -\frac{\Gamma_C(\delta\omega_C)^2}{2\pi} e^{-i(\omega_C-\omega_A)(t-t')} \int_{-\infty}^{\infty} d\omega \frac{e^{-i(\omega-\omega_C)(t-t')}}{(\omega-\omega_C)^2 + (\delta\omega_C)^2} \\ &= -\frac{1}{2}\Gamma_C\delta\omega_C e^{-i(\omega_C-\omega_A)(t-t')} e^{-\delta\omega_C|t-t'|}, \end{aligned} \quad (6.52)$$

and thus the integral equation (6.30) corresponds to the differential equation [101]

$$\ddot{C}_u(t) + [i(\omega_C - \omega_A) + \delta\omega_C] \dot{C}_u(t) + \frac{1}{2}\Gamma_C\delta\omega_C C_u(t) = 0. \quad (6.53)$$

where Γ_C is the decay rate at the cavity resonance [i.e., $\omega_A = \omega_C$ in the rate formulas (6.48) and (6.51)], and $\delta\omega_C$ is the width of the cavity resonance. For small absorption,

$\gamma \ll \omega_T, \omega_P, \omega_P^2/\omega_T$, the resonance lines in the band-gap zone can be regarded as being Lorentzians, and

$$\delta\omega_C = \frac{c\Gamma_0}{R_2\Gamma_C}. \quad (6.54)$$

Equation (6.53) reveals that (under the assumptions made) the upper-state probability amplitude of the atom obeys the equation of motion for a damped harmonic oscillator. In the strong-coupling regime, where $\omega_A = \omega_C$ and $\Gamma_C \gg \delta\omega_C$, damped Rabi oscillations are observed:²⁵

$$|C_u(t)|^2 = e^{-\delta\omega_C t} \cos^2(\Omega t/2), \quad (6.55)$$

where the Rabi frequency reads

$$\Omega = \sqrt{2\Gamma_C \delta\omega_C}. \quad (6.56)$$

Typical examples for the time evolution of the upper-state occupation probability

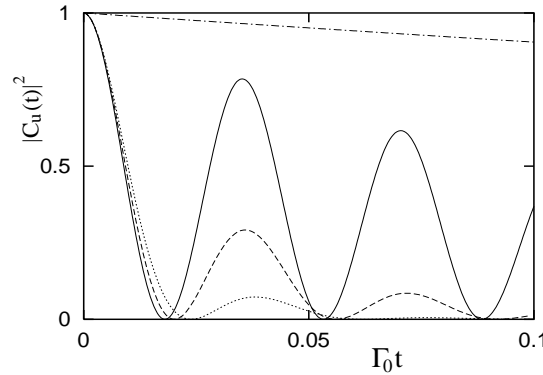


Figure 7: The temporal evolution of the occupation probability $|C_u(t)|^2$ of the upper atomic state is shown for $R_2 = 30\lambda_T$, $R_1 - R_2 = \lambda_T$, $\omega_P = 0.5\omega_T$, $\omega_A = 1.046448\omega_T$, $\Gamma_0\lambda_T/(2c) = 10^{-6}$, and $\gamma = 10^{-4}\omega_T$ (solid line), $\gamma = 5 \times 10^{-4}\omega_T$ (dashed line), $\gamma = 10^{-3}\omega_T$ (dotted line). For comparison, the exponential decay in free space is shown (dashed-dotted line).

are shown in Fig. 7. The curves are the exact (numerical) solutions of the integral equation (6.30) [together with the kernel function (6.31)] for the model permittivity (6.50). The figure shows that with increasing value of the intrinsic damping constant γ of the material of the cavity wall the Rabi oscillations become less pronounced. Physically, larger γ means larger absorption probability of the emitted photon by the cavity wall and thus reduced probability of atom-field energy interchange.

²⁵Note that in the opposite case where $\Gamma_C \ll \delta\omega_C$ the solution of Eq. (6.53) is $|C_u(t)|^2 = e^{-\Gamma_C t}$ for $\omega_A = \omega_C$.

7 Extensions to other media

So far we have focused our attention to quantum electrodynamics in absorbing, isotropic, non-magnetic, linear media. The quantization scheme outlined in Sec. 3 can, of course, also be generalized to other media. The starting point is the basic formula (3.35) relating the operator of the electric field strength to the dynamical variables of the system composed of the electromagnetic field and the medium. Recalling Eqs. (3.16) and (3.31), we may write Eq. (3.35) as

$$\hat{\mathbf{E}}(\mathbf{r}, \omega) = i\omega\mu_0 \int d^3\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}', \omega), \quad (7.1)$$

where

$$\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega) = \omega \sqrt{\frac{\hbar\varepsilon_0}{\pi}} \varepsilon_{\mathbf{I}}(\mathbf{r}, \omega) \hat{\mathbf{f}}(\mathbf{r}, \omega). \quad (7.2)$$

In what follows we briefly comment on possible extensions.

7.1 Amplifying media

Let us consider a dielectric which in some space and frequency regions is amplifying, but in other ones absorbing. Amplification can be described by a complex permittivity $\varepsilon(\mathbf{r}, \omega)$ which exhibits the familiar properties as in the case of absorption, most importantly, it still fulfills the Kramers–Kronig relations (3.19) and (3.20), except that the imaginary part is negative, $\varepsilon_{\mathbf{I}}(\mathbf{r}, \omega) < 0$. It can be shown that the quantization scheme in Sec. 3 also applies to amplifying media if Eq. (7.2) is modified according to [40]

$$\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega) = \omega \sqrt{\frac{\hbar\varepsilon_0}{\pi} |\varepsilon_{\mathbf{I}}(\mathbf{r}, \omega)|} \left[\Theta(\varepsilon_{\mathbf{I}}) \hat{\mathbf{f}}(\mathbf{r}, \omega) + \Theta(-\varepsilon_{\mathbf{I}}) \hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega) \right] \quad (7.3)$$

[$\Theta(x)$, Heaviside step function], which reflects the well-known fact that amplification requires the roles of the noise annihilation and creation operators to be exchanged [121, 122, 123, 124, 125]. Substitution of this expression into Eq. (7.1) yields the representation of the (ω -components of the) operator of the electric field strength in terms of the fundamental fields, which replaces Eq. (3.35). All the other formulas in Sec. 3 remain valid. In particular, the proof of the commutation relations (3.41) and (3.42) can be given in the same way as in Appendix B for absorbing media.

7.2 Anisotropic media

In anisotropic dielectrics the scalar $\varepsilon(\mathbf{r}, \omega)$ has to be replaced by a tensor $\boldsymbol{\varepsilon}(\mathbf{r}, \omega)$, which is symmetric for reciprocal media (see [58]),

$$\varepsilon_{ij}(\mathbf{r}, \omega) = \varepsilon_{ji}(\mathbf{r}, \omega). \quad (7.4)$$

Thus the Green tensor has to be determined from the partial differential equation

$$\nabla \times \nabla \times \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2}{c^2} \boldsymbol{\varepsilon}(\mathbf{r}, \omega) \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \boldsymbol{\delta}(\mathbf{r} - \mathbf{r}'), \quad (7.5)$$

which replaces Eq. (3.24). The solution for a uniaxial bulk dielectric is given in Eq. (A.39). The symmetry property (7.4) ensures that the reciprocity relation (3.27) is valid, and the generalization of Eq. (3.28) reads

$$\int d^3\mathbf{s} \frac{\omega^2}{c^2} \varepsilon_{ikl}(\mathbf{s}, \omega) G_{ik}(\mathbf{s}, \mathbf{r}, \omega) G_{jl}^*(\mathbf{s}, \mathbf{r}', \omega) = \text{Im } G_{ij}(\mathbf{r}, \mathbf{r}', \omega). \quad (7.6)$$

The extension of Eq. (7.2) to anisotropic, absorbing media then reads²⁶

$$\hat{\mathbf{j}}_{\mathbf{N}}(\mathbf{r}, \omega) = \omega \sqrt{\frac{\hbar \varepsilon_0}{\pi}} \varepsilon_{\mathbf{I}}^{1/2}(\mathbf{r}, \omega) \hat{\mathbf{f}}(\mathbf{r}, \omega), \quad (7.7)$$

where

$$\varepsilon_{\mathbf{I}}^{1/2}(\mathbf{r}, \omega) = \mathbf{O}(\mathbf{r}, \omega) \varepsilon_{\mathbf{I}}'^{1/2}(\mathbf{r}, \omega) \mathbf{O}^{-1}(\mathbf{r}, \omega). \quad (7.8)$$

Here the orthogonal matrix $\mathbf{O}(\mathbf{r}, \omega)$ transforms $\varepsilon_{\mathbf{I}}(\mathbf{r}, \omega)$ into its (positive) diagonal form $\varepsilon_{\mathbf{I}}'(\mathbf{r}, \omega)$,²⁷

$$\varepsilon_{\mathbf{I}}'(\mathbf{r}, \omega) = \mathbf{O}^{-1}(\mathbf{r}, \omega) \varepsilon_{\mathbf{I}}(\mathbf{r}, \omega) \mathbf{O}(\mathbf{r}, \omega). \quad (7.9)$$

Substitution of Eq. (7.7) into Eq. (7.1) yields the (ω -components of the) operator of the electric field strength expressed in terms of the fundamental fields, which replaces Eq. (3.35). The further procedure is same as outlined in Sec. 3, and it can again be shown that the commutation relations (3.41) and (3.42) are fulfilled.

7.3 Magnetic media

Let us now include a possible magnetic response of the matter in the quantization scheme, restricting our attention to isotropic media, so that the susceptibility $\mu(\mathbf{r}, \omega)$ is a scalar. We start from the constitutive relations, written in a non-standard form, which preserves the interpretation of the fields \mathbf{E} and \mathbf{B} as the fundamental electro-magnetic fields, i.e.,

$$\hat{\mathbf{D}}(\mathbf{r}, \omega) = \varepsilon_0 \hat{\mathbf{E}}(\mathbf{r}, \omega) + \hat{\mathbf{P}}(\mathbf{r}, \omega), \quad (7.10)$$

$$\hat{\mathbf{H}}(\mathbf{r}, \omega) = \kappa_0 \hat{\mathbf{B}}(\mathbf{r}, \omega) - \hat{\mathbf{M}}(\mathbf{r}, \omega). \quad (7.11)$$

Here, the polarization is given according to Eq. (3.9),

$$\hat{\mathbf{P}}(\mathbf{r}, \omega) = \varepsilon_0 [\varepsilon(\mathbf{r}, \omega) - 1] \hat{\mathbf{E}}(\mathbf{r}, \omega) + \hat{\mathbf{P}}_{\mathbf{N}}(\mathbf{r}, \omega), \quad (7.12)$$

and the magnetization reads

$$\hat{\mathbf{M}}(\mathbf{r}, \omega) = \kappa_0 [1 - \kappa(\mathbf{r}, \omega)] \hat{\mathbf{B}}(\mathbf{r}, \omega) + \hat{\mathbf{M}}_{\mathbf{N}}(\mathbf{r}, \omega), \quad (7.13)$$

where $\kappa(\mathbf{r}, \omega) = \mu^{-1}(\mathbf{r}, \omega)$ and $\kappa_0 = \mu_0^{-1}$. It is not difficult to prove that the Green tensor now obeys the equation

$$\left[\nabla \times \kappa(\mathbf{r}, \omega) \nabla \times - \frac{\omega^2}{c^2} \varepsilon(\mathbf{r}, \omega) \right] \mathbf{G}(\mathbf{r}, \mathbf{s}, \omega) = \delta(\mathbf{r} - \mathbf{s}). \quad (7.14)$$

²⁶Using the results of Sec. 7.1, it is not difficult to include in the scheme also amplification.

²⁷Note that, depending on the type of anisotropy in the dielectric, the direction of the optical axes might be frequency dependent [126].

Subsequently, the integral relation (3.28) has to be replaced by

$$\begin{aligned} & \int d^3\mathbf{s} \kappa_I(\mathbf{s}, \omega) \partial_n^s G_{ki}(\mathbf{s}, \mathbf{r}, \omega) [\partial_k^s G_{nj}^*(\mathbf{s}, \mathbf{r}', \omega) - \partial_n^s G_{kj}^*(\mathbf{s}, \mathbf{r}', \omega)] \\ & + \int d^3\mathbf{s} \frac{\omega^2}{c^2} \varepsilon_I(\mathbf{s}, \omega) G_{in}(\mathbf{s}, \mathbf{r}, \omega) G_{jn}^*(\mathbf{s}, \mathbf{r}', \omega) = \text{Im } G_{ij}(\mathbf{r}, \mathbf{r}', \omega), \end{aligned} \quad (7.15)$$

which can be proved correct in analogy to the derivation given in Appendix A for non-magnetic matter.

The contribution to $\hat{\mathbf{j}}_N(\mathbf{r}, \omega)$ of the “magnetization” changes Eq. (7.2) to²⁸

$$\hat{\mathbf{j}}_N(\mathbf{r}, \omega) = \omega \sqrt{\frac{\hbar \varepsilon_0}{\pi}} \varepsilon_I(\mathbf{r}, \omega) \hat{\mathbf{f}}_e(\mathbf{r}, \omega) + \nabla \times \sqrt{\frac{\hbar \kappa_0}{\pi}} |\kappa_I(\mathbf{r}, \omega)| \hat{\mathbf{f}}_m(\mathbf{r}, \omega), \quad (7.16)$$

with $\hat{\mathbf{f}}_e(\mathbf{r}, \omega)$ and $\hat{\mathbf{f}}_m(\mathbf{r}, \omega)$ being the bosonic vector fields associated with the electric and magnetic properties respectively. Here we have assumed that the medium is absorbing, so that $\mu_I(\mathbf{r}, \omega) > 0$ [51] and thus $\kappa_I(\mathbf{r}, \omega) = -\mu_I(\mathbf{r}, \omega)/|\mu(\mathbf{r}, \omega)|^2 < 0$.

Substituting for $\hat{\mathbf{j}}_N(\mathbf{r}', \omega)$ in Eq. (7.1) the result of Eq. (7.16), we obtain the (ω -components of the) operator of the electric field strength $\hat{\mathbf{E}}(\mathbf{r}, \omega)$ expressed in terms of the dynamical variables of the system. The fields $\hat{\mathbf{B}}(\mathbf{r}, \omega)$ and $\hat{\mathbf{D}}(\mathbf{r}, \omega)$ can then be constructed in the same way as in Sec. 3, and by means of Eq. (7.11) the field $\hat{\mathbf{H}}(\mathbf{r}, \omega)$ can be obtained accordingly. As before, the commutation relations (3.41) and (3.42) are fulfilled.

7.4 Nonlinear media

In general, the linear response of a dielectric medium to the electromagnetic field is only the first term in the series expansion of the polarization in powers of the electric field strength, and in strong fields nonlinear interactions play an important role. For example, the propagating of intense light in Kerr-type nonlinear media can lead to the formation of soliton-like pulses, the dispersion of the group velocity being compensated by the Kerr nonlinearity [127, 128, 129]. There have been a number of approaches to quantize the electromagnetic field in nonlinear dielectrics [24, 25, 26, 27], but mostly absorption is not taken into account in a consistent way.

A possible way to treat absorption is to include in the Hamiltonian (3.34) an appropriately chosen nonlinear term that may be thought of as being an integral over a nonlinear function of the medium polarization field $\mathbf{P}(\mathbf{r})$, and thus the extended Hamiltonian is given by

$$\hat{H} = \int d^3\mathbf{r} \int_0^\infty d\omega \hbar \omega \hat{\mathbf{f}}^\dagger(\mathbf{r}, \omega) \hat{\mathbf{f}}(\mathbf{r}, \omega) + \hat{H}_{\text{NL}} \quad (7.17)$$

with

$$\hat{H}_{\text{NL}} = \int d^3\mathbf{r} h_{\text{NL}}[\hat{\mathbf{P}}(\mathbf{r})], \quad (7.18)$$

²⁸Note that Eq.(3.16) changes to $\hat{\mathbf{j}}_N(\mathbf{r}, \omega) = -i\omega \hat{\mathbf{P}}_N(\mathbf{r}, \omega) + \nabla \times \hat{\mathbf{M}}_N(\mathbf{r}, \omega)$.

where

$$\hat{\mathbf{P}}(\mathbf{r}) = \int d\omega \hat{\mathbf{P}}(\mathbf{r}, \omega) + \text{H.c.} \quad (7.19)$$

with $\hat{\mathbf{P}}(\mathbf{r}, \omega)$ being defined according to Eq. (3.9).

The Heisenberg equation of motion of $\hat{\mathbf{f}}(\mathbf{r}, \omega)$ then reads

$$i\hbar \dot{\hat{\mathbf{f}}}(\mathbf{r}, \omega) = [\hat{\mathbf{f}}(\mathbf{r}, \omega), \hat{H}] = \hbar\omega \hat{\mathbf{f}}(\mathbf{r}, \omega) + [\hat{\mathbf{f}}(\mathbf{r}, \omega), \hat{H}_{\text{NL}}], \quad (7.20)$$

which can be rewritten as $\hat{\omega}\hat{\mathbf{f}}(\mathbf{r}, \omega) = \omega\hat{\mathbf{f}}(\mathbf{r}, \omega)$, where

$$\hat{\omega} = i\partial_t + \hbar^{-1}\hat{H}_{\text{NL}}^\times, \quad (7.21)$$

with $\hat{H}_{\text{NL}}^\times \hat{O} \equiv [\hat{H}_{\text{NL}}, \hat{O}]$. Hence the frequency-integrated (operator version of) Eq. (3.22) changes to [49]

$$\nabla \times \nabla \times \hat{\mathbf{E}}(\mathbf{r}) - \hat{K}(\mathbf{r}) \hat{\mathbf{E}}(\mathbf{r}) = \int d\omega i\omega\mu_0 \hat{\mathbf{j}}_{\text{N}}(\mathbf{r}, \omega) + \text{H.c.}, \quad (7.22)$$

with \hat{K} being the superoperator

$$\hat{K}(\mathbf{r}) = \frac{1}{c^2} \left(i\partial_t + \hbar^{-1}\hat{H}_{\text{NL}}^\times \right)^2 \varepsilon(\mathbf{r}, i\partial_t + \hbar^{-1}\hat{H}_{\text{NL}}^\times). \quad (7.23)$$

A similar equation holds for the (transverse) vector potential, from which the $\hat{\mathbf{B}}$ -field can be obtained. Obviously, the approach ensures that the commutation relations (3.41) and (3.42) are satisfied.

For a linear medium, the superoperator \hat{K} simply describes the effects of dispersion and absorption, but for a nonlinear medium it introduces via $\hat{H}_{\text{NL}}^\times$ additional nonlinear coupling terms. An application of Eq. (7.22) to one-dimensional propagation of light in a Kerr-type nonlinear medium is considered in [49], and a simplified version has been used to study soliton propagation in absorbing optical fibres [130].

Acknowledgements We are grateful to Ho Trung Dung, Tomáš Opatrný, Eduard Schmidt, and Adriaan Tip for valuable discussions and comments. This work was supported by the Deutsche Forschungsgemeinschaft.

References

- [1] J.M. Jauch and K.M. Watson, Phys. Rev. **74**, 950 (1948); *ibid.* **74**, 1485 (1948).
- [2] Y.R. Shen, Phys. Rev. **155**, 921 (1967).
- [3] C.K. Carniglia and L. Mandel, Phys. Rev. D **3**, 280 (1971).
- [4] I. Bialynicki-Birula and J.B. Brojan, Phys. Rev. D **5**, 485 (1972).
- [5] I. Abram, Phys. Rev. A **35**, 4661 (1987).
- [6] Z. Bialynicki-Birula and I. Bialynicki-Birula, J. Opt. Soc. Am. B **4**, 1621 (1987).
- [7] L. Knöll, W. Vogel, and D.-G. Welsch, Phys. Rev. A **36**, 3803 (1987).
- [8] T.A.B. Kennedy and E.M. Wright, Phys. Rev. A **38**, 212 (1988).
- [9] H. Khosravi and R. Loudon, Proc. R. Soc. London Ser. A **433**, 337 (1991); *ibid.* **436**, 373 (1992).
- [10] R.J. Glauber and M. Lewenstein, Phys. Rev. A **43**, 467 (1991).
- [11] L. Knöll and D.-G. Welsch, Prog. Quant. Electr. **16**, 135 (1992).
- [12] W. Vogel and D.-G. Welsch, *Lectures on Quantum Optics* (Akademie Verlag, Berlin, 1994).
- [13] B.J. Dalton, E.S. Guerra, and P.L. Knight, Phys. Rev. A **54**, 2292 (1996).
- [14] M. Bordag, K. Kirsten, and D.V. Vassilevich, J. Phys. A: Math. Gen. **31**, 2381 (1998).
- [15] B.J. Dalton, S.M. Barnett, and P.L. Knight, J. Mod. Opt. **46**, 1315 (1999); *ibid.* **46**, 1495 (1999).
- [16] K.M. Watson and J.M. Jauch, Phys. Rev. **75**, 1249 (1949).
- [17] G.S. Agarwal, Phys. Rev. A **11**, 230 (1975), Phys. Rev. A **11**, 243 (1975), Phys. Rev. A **11**, 253 (1975), Phys. Rev. A **12**, 1475 (1975).
- [18] B. Huttner, J.J. Baumberg, and S.M. Barnett, Europhys. Lett. **16**, 177 (1991).
- [19] P.D. Drummond, Phys. Rev. A **42**, 6845 (1991).
- [20] P.W. Milonni, J. Mod. Opt. **42**, 1991 (1995).
- [21] D.J. Santos and R. Loudon, Phys. Rev. A **52**, 1538 (1995).
- [22] Z. Hradil, Phys. Rev. A **53**, 3687 (1996).

- [23] P.D. Drummond and M. Hillery, Phys. Rev. A **59**, 691 (1999).
- [24] M. Hillery and L.D. Mlodinow, Phys. Rev. A **30**, 1860 (1984).
- [25] I. Abram and E. Cohen, Phys. Rev. A **44**, 500 (1991).
- [26] L.G. Joneckis and J.H. Shapiro, J. Opt. Soc. Am. B **10**, 1102 (1993).
- [27] Lu-Ming Duan and Guang-Can Guo, Phys. Rev. A **56**, 925 (1997).
- [28] M. Fleischhauer and M. Schubert, J. Mod. Opt. **38**, 677 (1991).
- [29] L. Knöll and U. Leonhardt, J. Mod. Opt. **39**, 1253 (1992).
- [30] B. Huttner and S.M. Barnett, Europhys. Lett. **18**, 487 (1992), Phys. Rev. A **46**, 4302 (1992).
- [31] S.-T. Ho and P. Kumar, J. Opt. Soc. Am. B **10**, 1620 (1993).
- [32] R. Matloob, R. Loudon, S.M. Barnett, and J. Jeffers, Phys. Rev. A **52**, 4823 (1995).
- [33] T. Gruner and D.-G. Welsch, Phys. Rev. A **53**, 1818 (1996).
- [34] T. Gruner and D.-G. Welsch, *Third Workshop on Quantum Field Theory under the Influence of External Conditions* (Leipzig, 1995) [B.G. Teubner Verlagsgesellschaft, Stuttgart-Leipzig, 1996].
- [35] S.M. Barnett, C.R. Gilson, B. Huttner, and N. Imoto, Phys. Rev. Lett. **77**, 1739 (1996).
- [36] R. Matloob and R. Loudon, Phys. Rev. A **53**, 4567 (1996).
- [37] A. Tip, Phys. Rev. A **56**, 5022 (1997).
- [38] A. Tip, Phys. Rev. A **57**, 4818 (1998).
- [39] Ho Trung Dung, L. Knöll, and D.-G. Welsch, Phys. Rev. A **57**, 3931 (1998).
- [40] S. Scheel, L. Knöll, and D.-G. Welsch, Phys. Rev. A **58**, 700 (1998).
- [41] J.R. Anglin and W.H. Zurek, Phys. Rev. D **53**, 7327 (1996).
- [42] A. Bechler, J. Mod. Opt. **46**, 901 (1999).
- [43] R. Matloob, Phys. Rev. A **60**, 50 (1999).
- [44] O. DiStefano, S. Savasta, and R. Girlanda, Phys. Rev. A **60**, 1614 (1999).
- [45] O. DiStefano, S. Savasta, and R. Girlanda, Phys. Rev. A **61**, 023803 (2000).
- [46] J.J. Hopfield, Phys. Rev. **112**, 1555 (1958).

- [47] U. Fano, Phys. Rev. **103**, 1202 (1956).
- [48] T. Gruner and D.-G. Welsch, Phys. Rev. A **51**, 3246 (1995).
- [49] E. Schmidt, J. Jeffers, S.M. Barnett, L. Knöll, and D.-G. Welsch, J. Mod. Opt. **45**, 377 (1997).
- [50] M. Altarelli, D.L. Dexter, H.M. Nussenzweig, and D.Y. Smith, Phys. Rev. B **6**, 4502 (1972).
- [51] L.D. Landau and E.M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon Press, 1960).
- [52] A.A. Abrikosov, L.P. Gorkov, and I.E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics* (Dover, New York, 1975).
- [53] E.A. Power and S. Zienau, Phil. Trans. Roy. Soc. A**251**, 427 (1959).
- [54] D.P. Craig and T. Thirunamachandran, *Molecular Quantum Electrodynamics* (Academic Press, London, 1984).
- [55] J.R. Ackerhalt and P.W. Milonni, J. Opt. Soc. Am. B **1**, 116 (1984).
- [56] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Photons and Atoms, Introduction to Quantum Electrodynamics* (Wiley, New York, 1989).
- [57] T. Gruner and D.-G. Welsch, Phys. Rev. A **54**, 1661 (1996).
- [58] W.C. Chew, *Waves and Fields in Inhomogeneous Media* (IEEE Press, New York, 1995).
- [59] M. Patra and C.W.J. Beenakker, Phys. Rev. A. **61**, 063805 (2000).
- [60] B. Yurke, S.L. McCall, and J.R. Klauder, Phys. Rev. A **33**, 4033 (1986).
- [61] S. Prasad, M.O. Scully, and W. Martienssen, Opt. Commun. **62**, 139 (1987).
- [62] Z.Y. Ou, C.K. Hong, and L. Mandel, Opt. Commun. **63**, 118 (1987).
- [63] H. Fearn and R. Loudon, Opt. Commun. **64**, 485 (1987).
- [64] R.A. Campos, B.E.A. Saleh, and M.C. Teich, Phys. Rev. A **40**, 1371 (1989).
- [65] U. Leonhardt, Phys. Rev. A **48**, 3265 (1993).
- [66] L. Knöll, S. Scheel, E. Schmidt, D.-G. Welsch, and A.V. Chizhov, Phys. Rev. A **59**, 4716 (1999).
- [67] D. Giulini, E. Joos, C. Kiefer, J. Kupsch, I.-O. Stamatescu, and H.D. Zeh, *Decoherence and the Appearance of a Classical World in Quantum Theory* (Springer, Heidelberg, 1996).

- [68] K.E. Cahill and R.J. Glauber, Phys. Rev. **177**, 1857 (1969); *ibid.* **177**, 1882 (1969).
- [69] J. Peřina, *Quantum Statistics of Linear and Nonlinear Optical Phenomena* (Kluwer Academic Publishers, Dordrecht/Boston/London, 1991)
- [70] S. Scheel, L. Knöll, T. Opatrny, and D.-G. Welsch, Phys. Rev. A **62**, 043803 (2000).
- [71] C.W. Gardiner, *Quantum Noise* (Springer, Berlin, 1991).
- [72] C.H. Bennett, G. Brassard, C. Crepeau, R. Josza, A. Peres, and W.K. Wootters, Phys. Rev. Lett. **70**, 1895 (1993).
- [73] S.L. Braunstein and H.J. Kimble, Phys. Rev. Lett. **80**, 869 (1998).
- [74] A. Furusawa, J.L. Sørensen, S.L. Braunstein, C.A. Fuchs, H.J. Kimble, and E.S. Polzik, Science **282**, 706 (1998).
- [75] C.H. Bennett and G. Brassard, *Proceedings of IEEE Conference on Computers, Systems and Signal Processing*, Bangalore, 175 (1984).
- [76] A. Peres, *Quantum Theory: Concepts and Methods* (McGraw-Hill, London, 1984).
- [77] V. Vedral and M.B. Plenio, Phys. Rev. A **57**, 1619 (1998).
- [78] M. Horodecki, P. Horodecki, and R. Horodecki, Phys. Rev. Lett. **84**, 2014 (2000).
- [79] S. Scheel, L. Knöll, T. Opatrny, and D.-G. Welsch, Proceedings of the 7th Central-European Workshop on Quantum Optics, Balatonfüred, Hungary, April 28 – May 1, 2000, acta physica slovacica **50**, 351 (2000); S. Scheel, T. Opatrny, and D.-G. Welsch, Paper presented at the International Conference on Quantum Optics 2000 and VIII Seminar on Quantum Optics Raubichi, BELARUS, May 28-31, 2000, arXiv: *quant-ph/0006026*.
- [80] T. Gruner and D.-G. Welsch, Opt. Commun. **134**, 447 (1997); *ibid.* **154**, 300 (1998).
- [81] A. Wehrl, Rev. Mod. Phys. **50**, 221 (1978).
- [82] M. Lewenstein and A. Sanpera, Phys. Rev. Lett. **80**, 2261 (1998).
- [83] A. Einstein, Z. Phys. **18**, 121 (1917).
- [84] J.M. Wylie and J.E. Sipe, Phys. Rev. A **30**, 1185 (1984).
- [85] D. Meschede, W. Jhe, and E.A. Hinds, Phys. Rev. A **41**, 1587 (1990).

- [86] S.M. Barnett, B. Huttner, and R. Loudon, Phys. Rev. Lett. **68**, 3698 (1992).
- [87] G. Juzeliūnas and D.L. Andrews, Phys. Rev. B **49**, 8751 (1994).
- [88] M. Fichet, F. Schuller, D. Bloch, and M. Ducloy, Phys. Rev. A **51**, 1553 (1995).
- [89] S.M. Barnett, B. Huttner, R. Loudon, and R. Matloob, J. Phys. B: At. Mol. Opt. Phys. **29**, 3763 (1996).
- [90] K. Koshino and A. Shimizu, Phys. Rev. A **53**, 4468 (1996).
- [91] M.S. Yeung and T.K. Gustafson, Phys. Rev. A **54**, 5227 (1996).
- [92] G. Juzeliūnas, Phys. Rev. A **55**, R4015 (1997).
- [93] M.S. Tomaš and Z. Lenac, Phys. Rev. A **56**, 4197 (1997).
- [94] S. Scheel, L. Knöll, D.-G. Welsch, and S.M. Barnett, Phys. Rev. A **60**, 1590 (1999).
- [95] S. Scheel, L. Knöll, and D.-G. Welsch, Phys. Rev. A **60**, 4094 (1999); *ibid.* **61**, 069901(E) (2000).
- [96] M. Fleischhauer and S. Yelin, Phys. Rev. A **59**, 2427 (1999).
- [97] M.S. Tomaš and Z. Lenac, Phys. Rev. A **60**, 2431 (1999).
- [98] M. Fleischhauer, Phys. Rev. A **60**, 2534 (1999).
- [99] P.W. Milonni, *The Quantum Vacuum: An Introduction to Quantum Electrodynamics* (Academic, San Diego, 1994).
- [100] R. Loudon, *Quantum Theory of Light* (Oxford University Press, 1983).
- [101] Ho Trung Dung, L. Knöll, and D.-G. Welsch, Phys. Rev. A **62**, 053804 (2000).
- [102] R.J. Cook and P.W. Milonni, Phys. Rev. A **35**, 5081 (1987).
- [103] X.P. Feng and K. Ujihara, IEEE J. Quantum Electron. **QE-25**, 2332 (1989).
- [104] Ho Trung Dung and K. Ujihara, Phys. Rev. A **60**, 4067 (1999).
- [105] C. Henkel and V. Sandoghdar, Opt. Commun. **158**, 250 (1998).
- [106] A.A. Maradudin and D.L. Mills, Phys. Rev. B **11**, 1392 (1975).
- [107] D.L. Mills and A.A. Maradudin, Phys. Rev. B **12**, 2943 (1975).
- [108] M.S. Tomaš, Phys. Rev. A **51**, 1347 (1995).

- [109] S. Scheel, L. Knöll, and D.-G. Welsch, Proceedings of the 6th Central-European Workshop on Quantum Optics, Château Chudobín, Czech Republic, April 30 – May 3, 1999, *acta physica slovacica* **49**, 585 (1999).
- [110] D.L. Dexter, *Phys. Rev.* **101**, 48 (1956).
- [111] B. di Bartolo, *Optical Interactions in Solids* (Wiley, New York, 1968).
- [112] A. Yariv, *Quantum Electronics* (Wiley, New York, 1975).
- [113] G. Nienhuis and C.Th.J. Alkemade, *Physica C* **81**, 181 (1976).
- [114] G.L.J.A. Rikken and Y.A.R.R. Kessener, *Phys. Rev. Lett.* **74**, 880 (1995).
- [115] F.J.P. Schuurmans, D.T.N. de Lang, G.H. Wegdam, R. Sprik, and A. Lagendijk, *Phys. Rev. Lett.* **80**, 5077 (1998).
- [116] P. de Vries and A. Lagendijk, *Phys. Rev. Lett.* **81**, 1381 (1998).
- [117] L.W. Li, P.S. Kooi, M.S. Leong, and T.S. Yeo, *IEEE Trans. Microwave Theory Tech.* **42**, 2302 (1994).
- [118] P.R. Berman (ed.), *Cavity Quantum Electrodynamics* (Academic, New York, 1994).
- [119] E.A. Hinds, in *Advances in Atomic, Molecular, and Optical Physics*, edited by D. Bates and B. Bederson (Academic, New York, 1991), Vol. 28.
- [120] M.I. Gorodetsky, A.A. Savchenkov, and V.S. Ilchenko, *Opt. Lett.* **21**, 453 (1996).
- [121] C.M. Caves, *Phys. Rev. D* **26**, 1817 (1982).
- [122] J. Jeffers, N. Imoto, and R. Loudon, *Phys. Rev. A* **47**, 3346 (1993).
- [123] J. Jeffers, S.M. Barnett, R. Loudon, R. Matloob, and M. Artoni, *Opt. Commun.* **131**, 66 (1996).
- [124] R. Matloob, R. Loudon, M. Artoni, S.M. Barnett, and J. Jeffers, *Phys. Rev. A* **55**, 1623 (1997).
- [125] M. Artoni and R. Loudon, *Phys. Rev. A* **57**, 622 (1998).
- [126] H. Römer, *Theoretische Optik* (VCH, Weinheim, 1994).
- [127] A. Hasegawa, *Optical solitons in fibers* (Springer, Berlin, 1989).
- [128] S.A. Akhmanov, V.A. Vysloukh, and A.S. Chirkin, *Optics of Femtosecond Laser Pulses* (AIP, New York, 1992).

- [129] J.R. Taylor (ed.), *Optical Solitons – Theory and Experiments* (Cambridge University Press, Cambridge, 1992).
- [130] E. Schmidt, L. Knöll, and D.-G. Welsch, Phys. Rev. A **59**, 2442 (1999).
- [131] R.G. Newton, *Scattering Theory of Waves and Particles*, (Springer, Berlin, 1982).
- [132] W.S. Weiglhofer, IEE Proc. H **137**, 5 (1990).

A The Green tensor

A.1 Basic relations

In Eq. (3.24), the Green tensor $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ as a function of \mathbf{r} and \mathbf{r}' can be regarded as being the matrix elements in the position basis of a (tensor-valued) Green operator $\hat{\mathbf{G}} = \hat{\mathbf{G}}(\omega)$ in an abstract 1-particle Hilbert space,

$$\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \langle \mathbf{r} | \hat{\mathbf{G}} | \mathbf{r}' \rangle. \quad (\text{A.1})$$

The matrix elements of the position operator $\hat{\mathbf{r}}$ are given by

$$\langle \mathbf{r} | \hat{\mathbf{r}} | \mathbf{r}' \rangle = \mathbf{r} \delta(\mathbf{r} - \mathbf{r}'), \quad (\text{A.2})$$

and the matrix elements of the associated momentum operator $\hat{\mathbf{p}}$,

$$[\hat{x}_i, \hat{p}_j] = i\delta_{ij}, \quad (\text{A.3})$$

read

$$\langle \mathbf{r} | \hat{\mathbf{p}} | \mathbf{r}' \rangle = \frac{1}{i} \nabla \delta(\mathbf{r} - \mathbf{r}'). \quad (\text{A.4})$$

Let $\hat{\mathbf{H}} = \hat{\mathbf{H}}(\omega)$ be the tensor-valued operator

$$\hat{\mathbf{H}} = i\hat{\mathbf{p}} \times \hat{\mathbf{p}} \times -\hat{q}^2 \hat{\mathbf{I}} = \hat{\mathbf{p}}^2 \hat{\mathbf{I}} - \hat{\mathbf{p}} \otimes \hat{\mathbf{p}} - \hat{q}^2 \hat{\mathbf{I}}, \quad (\text{A.5})$$

where

$$\hat{q}^2 = \frac{\omega^2}{c^2} \varepsilon(\hat{\mathbf{r}}, \omega), \quad (\text{A.6})$$

and $\hat{\mathbf{I}}$ is the unit operator,

$$\langle \mathbf{r} | \hat{\mathbf{I}} | \mathbf{r}' \rangle = \delta(\mathbf{r} - \mathbf{r}'). \quad (\text{A.7})$$

Equation (3.24) corresponds to the operator equation

$$\hat{\mathbf{H}} \hat{\mathbf{G}} = \hat{\mathbf{I}}, \quad (\text{A.8})$$

as can be easily seen. Writing down Eq. (A.8) in the position basis,

$$\int d^3\mathbf{s} \langle \mathbf{r} | \hat{\mathbf{H}} | \mathbf{s} \rangle \langle \mathbf{s} | \hat{\mathbf{G}} | \mathbf{r}' \rangle = \langle \mathbf{r} | \hat{\mathbf{I}} | \mathbf{r}' \rangle, \quad (\text{A.9})$$

and using Eqs. (A.2) and (A.4), we are just left with Eq. (3.24).

Now it is not difficult to prove Eq. (3.27). From Eq. (A.8) it follows that the equation

$$\hat{\mathbf{G}} = \hat{\mathbf{H}}^{-1} \quad (\text{A.10})$$

is valid, and thus we find, after multiplying it from the right by $\hat{\mathbf{H}}$,

$$\hat{\mathbf{G}}\hat{\mathbf{H}} = \hat{\mathbf{I}}, \quad (\text{A.11})$$

which in the position basis reads

$$\int d^3\mathbf{s} \langle \mathbf{r} | \hat{\mathbf{G}} | \mathbf{s} \rangle \langle \mathbf{s} | \hat{\mathbf{H}} | \mathbf{r}' \rangle = \langle \mathbf{r} | \hat{\mathbf{I}} | \mathbf{r}' \rangle. \quad (\text{A.12})$$

Using again Eqs. (A.2) and (A.4), after some straightforward calculation we derive

$$\left[\left(\partial_j^{r'} \partial_k^{r'} - \delta_{jk} \Delta^{r'} \right) - \delta_{jk} \frac{\omega^2}{c^2} \varepsilon(\mathbf{r}', \omega) \right] G_{ik}(\mathbf{r}, \mathbf{r}', \omega) = \delta_{ji} \delta(\mathbf{r}' - \mathbf{r}). \quad (\text{A.13})$$

Comparing Eq. (A.13) with Eq. (3.25), we immediately see the Green tensor indeed obeys Eq. (3.27).

In order to prove Eq. (3.28), we introduce operators $\hat{\mathbf{O}}^\dagger$ defined by

$$(\hat{\mathbf{O}}^\dagger)_{ij} = (\hat{\mathbf{O}}_{ji})^\dagger \equiv \hat{\mathbf{O}}_{ji}^\dagger. \quad (\text{A.14})$$

From Eq. (A.8) it then follows that

$$\hat{\mathbf{G}}^\dagger \hat{\mathbf{H}}^\dagger = \hat{\mathbf{I}}. \quad (\text{A.15})$$

Multiplying Eq. (A.8) from the left by $\hat{\mathbf{G}}^\dagger$ and Eq. (A.15) from the right by $\hat{\mathbf{G}}$ and subtracting the resulting equations from each other, we find

$$\hat{\mathbf{G}}^\dagger (\hat{\mathbf{H}} - \hat{\mathbf{H}}^\dagger) \hat{\mathbf{G}} = \hat{\mathbf{G}}^\dagger - \hat{\mathbf{G}}, \quad (\text{A.16})$$

which in Cartesian components reads

$$\hat{G}_{mi}^\dagger (\hat{H}_{mn} - \hat{H}_{nm}^\dagger) \hat{G}_{nj} = \hat{G}_{ji}^\dagger - \hat{G}_{ij}. \quad (\text{A.17})$$

From Eq. (A.5) it is easily seen that

$$\hat{H}_{mn} - \hat{H}_{nm}^\dagger = \delta_{mn} (\hat{q}^{2\dagger} - \hat{q}^2). \quad (\text{A.18})$$

Representing Eq. (A.17) [together with Eq. (A.18)] in the position basis and recalling the relations

$$\langle \mathbf{r} | \hat{G}_{ji}^\dagger | \mathbf{r}' \rangle = \langle \mathbf{r}' | \hat{G}_{ji} | \mathbf{r} \rangle^* = G_{ji}^*(\mathbf{r}', \mathbf{r}, \omega) = G_{ij}^*(\mathbf{r}, \mathbf{r}', \omega), \quad (\text{A.19})$$

we eventually arrive at Eq. (3.28).

A.1.1 Asymptotic behaviour

The Green tensor $G_{ij}(\mathbf{r}, \mathbf{r}', \omega)$ is holomorphic in the upper half-plane of complex ω , because of the holomorphic behaviour of $\varepsilon(\mathbf{r}, \omega)$. In order to study the behaviour of $G_{ij}(\mathbf{r}, \mathbf{r}', \omega)$ for $|\omega| \rightarrow \infty$ and $|\omega| \rightarrow 0$, we first introduce the tensor-valued projection operators

$$\hat{\mathbf{I}}^\perp = \hat{\mathbf{I}} - \hat{\mathbf{I}}^\parallel, \quad \hat{\mathbf{I}}^\parallel = \frac{\hat{\mathbf{p}} \otimes \hat{\mathbf{p}}}{\hat{\mathbf{p}}^2} \quad (\text{A.20})$$

and decompose $\hat{\mathbf{H}}$, Eq. (A.5), as

$$\hat{\mathbf{H}} = (\hat{\mathbf{p}}^2 - \hat{q}^2) \hat{\mathbf{I}}^\perp - \hat{q}^2 \hat{\mathbf{I}}^\parallel. \quad (\text{A.21})$$

Applying the Feshbach formula [131], we then may decompose the Green tensor operator $\hat{\mathbf{G}} = \hat{\mathbf{H}}^{-1}$, Eq. (A.10), as

$$\begin{aligned} \hat{\mathbf{G}} = \hat{\mathbf{H}}^{-1} &= \hat{\mathbf{I}}^\parallel (\hat{\mathbf{I}}^\parallel \hat{\mathbf{H}} \hat{\mathbf{I}}^\parallel)^{-1} \hat{\mathbf{I}}^\parallel \\ &+ [\hat{\mathbf{I}}^\perp - \hat{\mathbf{I}}^\parallel (\hat{\mathbf{I}}^\parallel \hat{\mathbf{H}} \hat{\mathbf{I}}^\parallel)^{-1} \hat{\mathbf{I}}^\parallel \hat{\mathbf{H}} \hat{\mathbf{I}}^\perp] \hat{\mathbf{K}} [\hat{\mathbf{I}}^\perp - \hat{\mathbf{I}}^\perp \hat{\mathbf{H}} \hat{\mathbf{I}}^\parallel (\hat{\mathbf{I}}^\parallel \hat{\mathbf{H}} \hat{\mathbf{I}}^\parallel)^{-1} \hat{\mathbf{I}}^\parallel], \end{aligned} \quad (\text{A.22})$$

where

$$\hat{\mathbf{K}} = [\hat{\mathbf{I}}^\perp \hat{\mathbf{H}} \hat{\mathbf{I}}^\perp - \hat{\mathbf{I}}^\perp \hat{\mathbf{H}} \hat{\mathbf{I}}^\parallel (\hat{\mathbf{I}}^\parallel \hat{\mathbf{H}} \hat{\mathbf{I}}^\parallel)^{-1} \hat{\mathbf{I}}^\parallel \hat{\mathbf{H}} \hat{\mathbf{I}}^\perp]^{-1}. \quad (\text{A.23})$$

It is not difficult to prove that

$$\hat{\mathbf{I}}^\parallel \hat{\mathbf{H}} \hat{\mathbf{I}}^\parallel = -\hat{\mathbf{I}}^\parallel \hat{q}^2 \hat{\mathbf{I}}^\parallel, \quad \hat{\mathbf{I}}^\parallel \hat{\mathbf{H}} \hat{\mathbf{I}}^\perp = -\hat{\mathbf{I}}^\parallel \hat{q}^2 \hat{\mathbf{I}}^\perp, \quad (\text{A.24})$$

$$\hat{\mathbf{I}}^\perp \hat{\mathbf{H}} \hat{\mathbf{I}}^\parallel = -\hat{\mathbf{I}}^\perp \hat{q}^2 \hat{\mathbf{I}}^\parallel, \quad \hat{\mathbf{I}}^\perp \hat{\mathbf{H}} \hat{\mathbf{I}}^\perp = \hat{\mathbf{I}}^\perp (\hat{\mathbf{p}}^2 - \hat{q}^2) \hat{\mathbf{I}}^\perp. \quad (\text{A.25})$$

Combining Eqs. (A.22) – (A.25) and using Eq. (A.6), we obtain for $\hat{\mathbf{G}}$ the expression

$$\begin{aligned} \hat{\mathbf{G}} &= -\frac{c^2}{\omega^2} \hat{\mathbf{I}}^\parallel (\hat{\mathbf{I}}^\parallel \hat{\varepsilon} \hat{\mathbf{I}}^\parallel)^{-1} \hat{\mathbf{I}}^\parallel \\ &+ [\hat{\mathbf{I}}^\perp - \hat{\mathbf{I}}^\parallel (\hat{\mathbf{I}}^\parallel \hat{\varepsilon} \hat{\mathbf{I}}^\parallel)^{-1} \hat{\mathbf{I}}^\parallel \hat{\varepsilon} \hat{\mathbf{I}}^\perp] \hat{\mathbf{K}} [\hat{\mathbf{I}}^\perp - \hat{\mathbf{I}}^\perp \hat{\varepsilon} \hat{\mathbf{I}}^\parallel (\hat{\mathbf{I}}^\parallel \hat{\varepsilon} \hat{\mathbf{I}}^\parallel)^{-1} \hat{\mathbf{I}}^\parallel], \end{aligned} \quad (\text{A.26})$$

where

$$\hat{\mathbf{K}} = \left[\hat{\mathbf{I}}^\perp \left(\hat{\mathbf{p}}^2 - \frac{\omega^2}{c^2} \hat{\varepsilon} \right) \hat{\mathbf{I}}^\perp + \frac{\omega^2}{c^2} \hat{\mathbf{I}}^\perp \hat{\varepsilon} \hat{\mathbf{I}}^\parallel (\hat{\mathbf{I}}^\parallel \hat{\varepsilon} \hat{\mathbf{I}}^\parallel)^{-1} \hat{\mathbf{I}}^\parallel \hat{\varepsilon} \hat{\mathbf{I}}^\perp \right]^{-1} \quad (\text{A.27})$$

$[\hat{\varepsilon} = \varepsilon(\hat{\mathbf{r}}, \omega)]$.

Now the desired limiting processes can be performed easily. For $|\omega| \rightarrow 0$ we find $[\hat{\varepsilon}^{(0)} = \varepsilon^{(0)}(\hat{\mathbf{r}}) = \varepsilon(\hat{\mathbf{r}}, \omega = 0)]$

$$\lim_{|\omega| \rightarrow 0} \frac{\omega^2}{c^2} \hat{\mathbf{G}} = -\hat{\mathbf{I}}^\parallel (\hat{\mathbf{I}}^\parallel \hat{\varepsilon}^{(0)} \hat{\mathbf{I}}^\parallel)^{-1} \hat{\mathbf{I}}^\parallel \quad (\text{A.28})$$

and

$$\lim_{|\omega| \rightarrow 0} \hat{q}^2 \hat{\mathbf{G}} = -\hat{\varepsilon}^{(0)} \hat{\mathbf{I}}^\parallel (\hat{\mathbf{I}}^\parallel \hat{\varepsilon}^{(0)} \hat{\mathbf{I}}^\parallel)^{-1} \hat{\mathbf{I}}^\parallel. \quad (\text{A.29})$$

For $|\omega| \rightarrow \infty$ we arrive at, on recalling that $\hat{\mathbf{I}}^{\parallel} \hat{\mathbf{I}}^{\perp} = \hat{\mathbf{I}}^{\perp} \hat{\mathbf{I}}^{\parallel} = 0$ and $\varepsilon(\mathbf{r}, \omega) \rightarrow 1$ if $|\omega| \rightarrow \infty$,

$$\lim_{|\omega| \rightarrow \infty} \frac{\omega^2}{c^2} \hat{\mathbf{G}} = \lim_{|\omega| \rightarrow \infty} \hat{q}^2 \hat{\mathbf{G}} = -\hat{\mathbf{I}}. \quad (\text{A.30})$$

Obviously, the first term on the right hand of Eq. (A.26) is the singular part of $\hat{\mathbf{G}}$ for $|\omega| \rightarrow 0$. Performing in that term the Taylor expansion

$$\varepsilon(\hat{\mathbf{r}}, \omega) = \varepsilon^{(0)}(\hat{\mathbf{r}}) + \omega \varepsilon^{(1)}(\hat{\mathbf{r}}) + \dots, \quad (\text{A.31})$$

where $\varepsilon^{(0)}(\hat{\mathbf{r}})$ is real and $\varepsilon^{(1)}(\hat{\mathbf{r}})$ is imaginary [see Eq. (3.21)], we find that

$$\text{Re } G_{ij}(\mathbf{r}, \mathbf{r}', \omega) \sim \omega^{-2} \quad (|\omega| \rightarrow 0) \quad (\text{A.32})$$

and

$$\text{Im } G_{ij}(\mathbf{r}, \mathbf{r}', \omega) \sim \omega^{-1} \quad (|\omega| \rightarrow 0). \quad (\text{A.33})$$

A.1.2 Isotropic bulk material

For a bulk material of given permittivity $\varepsilon(\omega)$ the Green tensor reads ($\boldsymbol{\rho} = \mathbf{r} - \mathbf{r}'$)

$$\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = [\nabla^r \otimes \nabla^r + \mathbf{I} q^2(\omega)] \frac{e^{iq(\omega)\rho}}{4\pi q^2(\omega)\rho}, \quad (\text{A.34})$$

where

$$q(\omega) = \sqrt{\varepsilon(\omega)} \frac{\omega}{c} = [n_{\text{R}}(\omega) + i n_{\text{I}}(\omega)] \frac{\omega}{c}. \quad (\text{A.35})$$

It can be split up into a longitudinal and a transverse part according to

$$\mathbf{G}^{\parallel}(\mathbf{r}, \mathbf{r}', \omega) = -\frac{1}{4\pi q^2} \left[\frac{4\pi}{3} \delta(\boldsymbol{\rho}) \mathbf{I} + \left(\mathbf{I} - \frac{3\boldsymbol{\rho} \otimes \boldsymbol{\rho}}{\rho^2} \right) \frac{1}{\rho^3} \right] \quad (\text{A.36})$$

and

$$\begin{aligned} \mathbf{G}^{\perp}(\mathbf{r}, \mathbf{r}', \omega) = & \frac{1}{4\pi q^2} \left\{ \left(\mathbf{I} - \frac{3\boldsymbol{\rho} \otimes \boldsymbol{\rho}}{\rho^2} \right) \frac{1}{\rho^3} \right. \\ & \left. + q^3 \left[\left(\frac{1}{q\rho} + \frac{i}{(q\rho)^2} - \frac{1}{(q\rho)^3} \right) \mathbf{I} - \left(\frac{1}{q\rho} + \frac{3i}{(q\rho)^2} - \frac{3}{(q\rho)^3} \right) \frac{\boldsymbol{\rho} \otimes \boldsymbol{\rho}}{\rho^2} \right] e^{iq\rho} \right\}. \end{aligned} \quad (\text{A.37})$$

In particular, from Eq. (A.37) it follows that

$$\text{Im } \mathbf{G}^{\perp}(\mathbf{r}, \mathbf{r}, \omega) = \lim_{\mathbf{r}' \rightarrow \mathbf{r}} \text{Im } \mathbf{G}^{\perp}(\mathbf{r}, \mathbf{r}', \omega) = \frac{\omega}{6\pi c} n_{\text{R}}(\omega) \mathbf{I}. \quad (\text{A.38})$$

A.1.3 Uniaxial bulk material

Following [132], the Green tensor for a homogeneous uniaxial dielectric can be given in the form of

$$\begin{aligned} \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = & q_t^{-2}(\omega) [\nabla^r \otimes \nabla^r + q_t^2(\omega) \varepsilon_c \boldsymbol{\varepsilon}^{-1}] \frac{e^{iq_t(\omega)\rho_e}}{4\pi\rho_e} \\ & - \left[\frac{\varepsilon_c e^{iq_t(\omega)\rho_e}}{\varepsilon_t} - \frac{e^{iq_t(\omega)\rho}}{4\pi\rho} \right] \frac{(\boldsymbol{\rho} \times \mathbf{c}) \otimes (\boldsymbol{\rho} \times \mathbf{c})}{(\boldsymbol{\rho} \times \mathbf{c})^2} \\ & - \left[\frac{e^{iq_t(\omega)\rho_e} - e^{iq_t(\omega)\rho}}{4\pi i q_t(\omega)} \right] \left[\frac{\mathbf{I} - \mathbf{c} \otimes \mathbf{c}}{(\boldsymbol{\rho} \times \mathbf{c})^2} - \frac{2(\boldsymbol{\rho} \times \mathbf{c}) \otimes (\boldsymbol{\rho} \times \mathbf{c})}{(\boldsymbol{\rho} \times \mathbf{c})^4} \right], \end{aligned} \quad (\text{A.39})$$

$[\rho_e^2 = \varepsilon_c \boldsymbol{\rho} \boldsymbol{\varepsilon}^{-1} \boldsymbol{\rho}, q_t = \sqrt{\varepsilon_t} \omega / c]$. Here, the vector \mathbf{c} is the unit vector parallel to the crystallographic axis of the medium, $\varepsilon_c = \varepsilon_c(\omega)$ the (complex) permittivity in that direction, and $\varepsilon_t = \varepsilon_t(\omega)$ the (complex) permittivity transverse to it. Note that for $\varepsilon_c = \varepsilon_t = \varepsilon$ [and $\boldsymbol{\varepsilon}^{-1} = \varepsilon^{-1} \mathbf{I}$] the Green tensor (A.34) for isotropic bulk material is recovered.

B Commutation relations

Let us first consider the commutation relations of the electric field and the vector potential. Using Eqs. (3.38) and (3.46) together with Eqs. (3.35) and (3.47), recalling the commutation relations (3.32) and (3.33), and applying the integral relation (3.28), after some algebra we derive

$$[\hat{E}_k(\mathbf{r}), \hat{E}_l(\mathbf{r}')] = 0, \quad (\text{B.1})$$

$$[\hat{A}_k(\mathbf{r}), \hat{A}_l(\mathbf{r}')] = 0, \quad (\text{B.2})$$

$$[\varepsilon_0 \hat{E}_k(\mathbf{r}), \hat{A}_l(\mathbf{r}')] = \int d^3\mathbf{s} \left[\frac{2i\hbar}{\pi} \int_0^\infty d\omega \frac{\omega}{c^2} \text{Im } G_{km}(\mathbf{r}, \mathbf{s}, \omega) \right] \delta_{ml}^\perp(\mathbf{s} - \mathbf{r}'). \quad (\text{B.3})$$

In Eq. (B.3) the ω -integral can be performed by applying the rule

$$\int_0^\infty d\omega \dots = \lim_{\epsilon \rightarrow 0} \int_\epsilon^\infty d\omega \dots \quad (\text{B.4})$$

Note that, according to Eq. (A.33), $\text{Im } G_{il}(\mathbf{r}, \mathbf{r}', \omega)$ behaves like ω^{-1} as ω approaches zero. Thus we may transform, on recalling Eq. (3.26), the ω -integral into a principal-part (\mathcal{P}) integral, so that Eq.(B.3) reads

$$[\varepsilon_0 \hat{E}_k(\mathbf{r}), \hat{A}_l(\mathbf{r}')] = \int d^3\mathbf{s} \left[\hbar \frac{\mathcal{P}}{\pi} \int_{-\infty}^\infty \frac{d\omega}{\omega} \frac{\omega^2}{c^2} G_{km}(\mathbf{r}, \mathbf{s}, \omega) \right] \delta_{ml}^\perp(\mathbf{s} - \mathbf{r}'). \quad (\text{B.5})$$

The evaluation of the ω -integral in Eq. (B.5) can be performed by means of contour-integral techniques. Since $G_{km}(\mathbf{r}, \mathbf{s}, \omega)$ is a holomorphic function of ω in

the upper complex frequency half-plane with the asymptotic behaviour according to Eq. (A.30), the ω -integrals can be calculated by contour integration along an infinitely small half-circle $|\omega| = \rho$, $\rho \rightarrow 0$, and an infinitely large half-circle $|\omega| = R$, $R \rightarrow \infty$, in the upper complex half-plane

$$\mathcal{P} \int_{-\infty}^{\infty} d\omega \dots = \lim_{\rho \rightarrow 0} \int_{\substack{|\omega|=\rho \\ \omega_I > 0}} d\omega \dots - \lim_{R \rightarrow \infty} \int_{\substack{|\omega|=R \\ \omega_I > 0}} d\omega \dots \quad (\text{B.6})$$

The definition of $\hat{\mathbf{I}}^{\perp(\parallel)}$ as given in Eq. (A.20) implies that

$$\langle \mathbf{r} | \hat{\mathbf{I}}^{\perp(\parallel)} | \mathbf{r}' \rangle = \boldsymbol{\delta}^{\perp(\parallel)}(\mathbf{r} - \mathbf{r}'), \quad (\text{B.7})$$

and thus we may write, on applying Eq. (A.1),

$$\int d^3\mathbf{s} \left[\int_{\mathcal{C}} \frac{d\omega}{\omega} \frac{\omega^2}{c^2} G_{km}(\mathbf{r}, \mathbf{s}, \omega) \right] \delta_{ml}^{\perp}(\mathbf{s} - \mathbf{r}') = \int_{\mathcal{C}} \frac{d\omega}{\omega} \frac{\omega^2}{c^2} \langle \mathbf{r} | \hat{\mathbf{G}}(\omega) \hat{\mathbf{I}}^{\perp} | \mathbf{r}' \rangle. \quad (\text{B.8})$$

Note that

$$\boldsymbol{\delta}^{\parallel}(\mathbf{r}) = -\boldsymbol{\nabla} \otimes \boldsymbol{\nabla} \frac{1}{4\pi|\mathbf{r}|} \quad (\text{B.9})$$

and

$$\boldsymbol{\delta}^{\perp}(\mathbf{r}) = \boldsymbol{\delta}(\mathbf{r}) - \boldsymbol{\delta}^{\parallel}(\mathbf{r}). \quad (\text{B.10})$$

From Eq. (A.28) it follows that

$$\lim_{|\omega| \rightarrow 0} \frac{\omega^2}{c^2} \hat{\mathbf{G}} \hat{\mathbf{I}}^{\perp} = -\hat{\mathbf{I}}^{\parallel} (\hat{\mathbf{I}}^{\parallel} \hat{\varepsilon}^{(0)} \hat{\mathbf{I}}^{\parallel})^{-1} \hat{\mathbf{I}}^{\parallel} \hat{\mathbf{I}}^{\perp} = 0 \quad (\text{B.11})$$

($\hat{\mathbf{I}}^{\parallel} \hat{\mathbf{I}}^{\perp} = 0$) and therefore the integral over the small half-circle vanishes. Finally, from Eq. (A.30) we see that

$$\lim_{|\omega| \rightarrow \infty} \frac{\omega^2}{c^2} \hat{\mathbf{G}} \hat{\mathbf{I}}^{\perp} = -\hat{\mathbf{I}}^{\perp}. \quad (\text{B.12})$$

Hence,

$$\begin{aligned} \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{\omega} \frac{\omega^2}{c^2} \langle \mathbf{r} | \hat{\mathbf{G}}(\omega) \hat{\mathbf{I}}^{\perp} | \mathbf{r}' \rangle \\ = 2i \int_0^{\infty} d\omega \frac{\omega}{c^2} \text{Im} \langle \mathbf{r} | \hat{\mathbf{G}}(\omega) \hat{\mathbf{I}}^{\perp} | \mathbf{r}' \rangle = i\pi \boldsymbol{\delta}^{\perp}(\mathbf{r} - \mathbf{r}'), \end{aligned} \quad (\text{B.13})$$

and the sought commutator reads

$$\left[\varepsilon_0 \hat{E}_k(\mathbf{r}), \hat{A}_l(\mathbf{r}') \right] = i\hbar \delta_{kl}^{\perp}(\mathbf{r} - \mathbf{r}'). \quad (\text{B.14})$$

Since the corresponding commutation relations for the displacement field [Eq. (3.40) together with Eqs. (3.37) and (3.35)] can be derived in a quite similar way, we renounce the derivation here and immediately give the result

$$\left[\hat{D}_k(\mathbf{r}), \hat{D}_l(\mathbf{r}') \right] = 0, \quad (\text{B.15})$$

$$\left[\hat{D}_k(\mathbf{r}), \hat{A}_l(\mathbf{r}') \right] = i\hbar \delta_{kl}^\perp(\mathbf{r} - \mathbf{r}'). \quad (\text{B.16})$$

This is the result we have expected, because the polarization $\hat{\mathbf{P}}(\mathbf{r}) = \hat{\mathbf{D}}(\mathbf{r}) - \varepsilon_0 \hat{\mathbf{E}}(\mathbf{r})$ is related to the degrees of freedom of the matter and it should therefore commute with the radiation field operators.

Recalling the relations $\nabla \times \hat{\mathbf{A}} = \hat{\mathbf{B}}$, $\hat{\Pi} = -\varepsilon_0 \hat{\mathbf{E}}^\perp$, $-\nabla \hat{\varphi} = \hat{\mathbf{E}}^\parallel$, and $\hat{\mathbf{P}} = \hat{\mathbf{D}} - \varepsilon_0 \hat{\mathbf{E}}$ and using the commutation relations (B.1), (B.2), (B.14), (B.15), and (B.16), we can then derive further commutation relations such as

$$\left[\varepsilon_0 \hat{E}_k(\mathbf{r}), \hat{B}_l(\mathbf{r}') \right] = -i\hbar \epsilon_{klm} \partial_m^r \delta(\mathbf{r} - \mathbf{r}'), \quad (\text{B.17})$$

$$\left[\hat{A}_k(\mathbf{r}), \hat{\Pi}_l(\mathbf{r}') \right] = i\hbar \delta_{kl}^\perp(\mathbf{r} - \mathbf{r}'), \quad (\text{B.18})$$

$$[\hat{\varphi}(\mathbf{r}), \hat{\varphi}(\mathbf{r}')] = [\hat{\varphi}(\mathbf{r}), \hat{A}_k(\mathbf{r}')] = [\hat{\varphi}(\mathbf{r}), \hat{E}_k(\mathbf{r}')] = 0, \quad (\text{B.19})$$

$$[\hat{\varphi}(\mathbf{r}), \hat{D}_k(\mathbf{r}')] = 0. \quad (\text{B.20})$$

It should be pointed out that the commutation relations given above are valid for both the medium-assisted electromagnetic field considered in Sec. 3 and the total electromagnetic field considered in Sec. 4. Needless to say that quantities of the medium-assisted electromagnetic field and quantities of the additional charged particles commute.

C Equations of motion

In order to prove that the quantization scheme yields the correct Maxwell equations and the Newtonian equations of motion of the charged particles, let us consider, e.g. the Maxwell equation (4.8). Using the minimal-coupling Hamiltonian (4.1), we find that

$$\begin{aligned} \dot{\hat{\mathbf{D}}}(\mathbf{r}) &= \frac{1}{i\hbar} [\hat{\mathbf{D}}(\mathbf{r}), \hat{H}] = \frac{1}{i\hbar} \int d^3\mathbf{r}' \int_0^\infty d\omega \hbar\omega [\hat{\mathbf{D}}(\mathbf{r}), \hat{\mathbf{f}}^\dagger(\mathbf{r}', \omega) \hat{\mathbf{f}}(\mathbf{r}', \omega)] \\ &\quad + \frac{1}{i\hbar} \sum_\alpha \frac{1}{2m_\alpha} [\hat{\mathbf{D}}(\mathbf{r}), [\hat{\mathbf{p}}_\alpha - q_\alpha \hat{\mathbf{A}}(\hat{\mathbf{r}}_\alpha)]^2] \\ &\quad + \frac{1}{2i\hbar} \int d^3\mathbf{r}' [\hat{\mathbf{D}}(\mathbf{r}), \hat{\rho}_A(\mathbf{r}') \hat{\varphi}_A(\mathbf{r}')] + \frac{1}{i\hbar} \int d^3\mathbf{r}' [\hat{\mathbf{D}}(\mathbf{r}), \hat{\rho}_A(\mathbf{r}') \hat{\varphi}_M(\mathbf{r}')]. \end{aligned} \quad (\text{C.1})$$

Recalling the definition (4.13) of the displacement field and applying the commutation relation (B.20), we may simplify Eq. (C.1) to

$$\begin{aligned} \dot{\hat{\mathbf{D}}}(\mathbf{r}) &= \frac{1}{i\hbar} \int d^3\mathbf{r}' \int_0^\infty d\omega \hbar\omega [\hat{\mathbf{D}}_M(\mathbf{r}), \hat{\mathbf{f}}^\dagger(\mathbf{r}', \omega) \hat{\mathbf{f}}(\mathbf{r}', \omega)] \\ &\quad + \frac{1}{i\hbar} \sum_\alpha \frac{1}{2m_\alpha} [\hat{\mathbf{D}}_M(\mathbf{r}), [\hat{\mathbf{p}}_\alpha - q_\alpha \hat{\mathbf{A}}(\hat{\mathbf{r}}_\alpha)]^2] \\ &\quad - \frac{\varepsilon_0}{i\hbar} \sum_\alpha \frac{1}{2m_\alpha} [\nabla \hat{\varphi}_A(\mathbf{r}), [\hat{\mathbf{p}}_\alpha - q_\alpha \hat{\mathbf{A}}(\hat{\mathbf{r}}_\alpha)]^2]. \end{aligned} \quad (\text{C.2})$$

Using Eqs. (3.39) and (3.40) together with Eqs. (3.36), (3.37), and (3.35) and recalling the basic commutation relations (3.32) and (3.33), we find that the first of the three commutators in Eq. (C.2) gives

$$\frac{1}{i\hbar} \int d^3\mathbf{r}' \int_0^\infty d\omega \hbar\omega [\hat{\mathbf{D}}_M(\mathbf{r}), \hat{\mathbf{f}}^\dagger(\mathbf{r}', \omega) \hat{\mathbf{f}}(\mathbf{r}', \omega)] = \frac{1}{\mu_0} \boldsymbol{\nabla} \times \hat{\mathbf{B}}(\mathbf{r}). \quad (\text{C.3})$$

In order to calculate the second commutator, we use the commutation relation (B.16) and obtain

$$\frac{1}{i\hbar} \sum_\alpha \frac{1}{2m_\alpha} [\hat{\mathbf{D}}_M(\mathbf{r}), [\hat{\mathbf{p}}_\alpha - q_\alpha \hat{\mathbf{A}}(\hat{\mathbf{r}}_\alpha)]^2] = -\hat{\mathbf{j}}_A^\perp(\mathbf{r}). \quad (\text{C.4})$$

Finally, recalling the definitions of $\hat{\varphi}_A(\mathbf{r})$ [Eq. (4.2)] and $\hat{\rho}_A(\mathbf{r})$ [Eq. (4.3)] and using the particle-operator commutation relations, we derive

$$-\frac{\varepsilon_0}{i\hbar} \sum_\alpha \frac{1}{2m_\alpha} [\boldsymbol{\nabla} \hat{\varphi}_A(\mathbf{r}), [\hat{\mathbf{p}}_\alpha - q_\alpha \hat{\mathbf{A}}(\hat{\mathbf{r}}_\alpha)]^2] = -\hat{\mathbf{j}}_A^\parallel(\mathbf{r}). \quad (\text{C.5})$$

In Eqs. (C.4) and (C.5), the transverse and longitudinal current densities $\hat{\mathbf{j}}_A^\perp(\mathbf{r})$ and $\hat{\mathbf{j}}_A^\parallel(\mathbf{r})$, respectively, are defined by

$$\hat{\mathbf{j}}_A^{\perp(\parallel)}(\mathbf{r}) = \frac{1}{2} \sum_\alpha q_\alpha \dot{\hat{\mathbf{r}}}_\alpha \boldsymbol{\delta}^{\perp(\parallel)}(\mathbf{r} - \hat{\mathbf{r}}_\alpha) + \text{H.c.}, \quad (\text{C.6})$$

where the velocity operator $\dot{\hat{\mathbf{r}}}_\alpha$ of the α th particle is

$$\dot{\hat{\mathbf{r}}}_\alpha = \frac{1}{i\hbar} [\hat{\mathbf{r}}_\alpha, \hat{H}] = \frac{1}{m_\alpha} [\hat{\mathbf{p}}_\alpha - q_\alpha \hat{\mathbf{A}}(\hat{\mathbf{r}}_\alpha)]. \quad (\text{C.7})$$

Substituting Eqs. (C.3), (C.4), and (C.5) back into Eq. (C.2), we just arrive at the Maxwell equation (4.8). The Maxwell equation (4.6) and the Newtonian equations of motion (4.10) and (4.11) can be derived in a quite similar way.